Advanced Light Source

Strategic Plan

2014–18

Table of Contents

Executive Summary

We know much about the structures of molecules and solids, yet despite all this knowledge, we still are not able to make the energy storage systems we need, develop efficient artificial photosynthesis, or form desired chemical bonds at will. Solving these and many other problems will involve deploying novel materials that function cooperatively in heterogeneous structures and environments. Research with soft x-ray beams will be an essential ingredient in developing and exploring these materials because we need to use the sharp shallow core levels, the next nearest energy levels to the valence levels we wish to probe, to determine “Where are the electrons?” and “How do they determine bonding, kinetics, magnetism, and many other properties?”

These questions are particularly important in materials that are intrinsically heterogeneous as well as in material devices with designed heterogeneity. For example, the properties of the wide-band-gap oxides $SrTiO_3$ and $LiAlO_3$ are well understood, yet a heterojunction between these two is metallic and even superconducting at low temperature. Similarly, a particular kinase enzyme will often appear in several different protein complexes, yet remarkably it functions differently in each assembly. Apparently, knowing the structure and properties of the components of a heterogeneous material often provides an incomplete picture of the resulting properties and function. We need tools both to uncover this heterogeneity as well as to measure the response of the components to perturbations on various spatial and temporal scales.

Developing predictive power for such emergent properties lies at the core of several of the Grand Challenges identified by the Department of Energy (DOE), Office of Basic Energy Sciences (BES), and will remain a primary focus of chemical, materials, and biological research for decades to come. How do we rationally build and understand chemical, material, and biological structures, and even structures that combine these three types of mater, to achieve a desired function? How do we endow such synthetic structures with feedback, regulation, self-assembly, and self-repair, which are essential ingredients of living systems, so that they maintain the desired function with high fidelity and efficiency over long periods of time? Just as an artificial photosynthetic cell will use a compartmentalized structure to convert solar energy directly to fuel, a “catalytic network” will assemble nanocatalytic centers into a mesoscale structure designed to perform a multistep chemical synthesis with high efficiency and selectivity at low temperature, a “transport network” will dynamically redirect the flow of energy to maximize energy storage or control energy conversion efficiency, and a “multifunctional oxide network” will amplify the sensitivity of these materials to applied fields to enable next-generation artificial neural networks.

The vision laid forth in this Advanced Light Source (ALS) Strategic Plan is to continue to develop and support cutting-edge soft x-ray tools for multiscale imaging of functional materials and mesoscale devices. The chemical and magnetic sensitivity of soft x-ray spectroscopy, combined with the spatial resolution of soft x-ray microscopies, provide a powerful suite of tools to probe heterogeneous materials and functional material devices to understand and help optimize their performance. In its first 20 years of operation, the ALS has led the world in developing these core soft x-ray capabilities, including in particular zone-plate focusing and advanced spectroscopies in diverse environments. The health of the innovative ALS culture is reflected by the ongoing design, construction, and commissioning of a "nanoARPES" capability to probe the electronic structure of mesoscale systems, a coherent soft x-ray scattering beamline to probe nanometer-scale chemical correlations eventually with nanosecond sensitivity, and a spin-resolved ARPES capability to understand and develop emerging ultralow-power spintronic materials and devices.

Though the ALS is nearly built out, its innovative culture will continue unabated well into the future by incorporating the latest technologies, by continually optimizing and updating existing capabilities, by seeking efficiencies wherever possible, and by repurposing existing beamlines to serve the most relevant science needs. The highest instrument priorities over the next few years will extend the capacity and capability of our soft x-ray tools to probe advanced energy materials and material structures. Most notably, we will install the Advanced Beamline for Energy Research (AMBER) to probe energy materials in diverse sample environments, we will develop and install a high-resolution resonant inelastic soft x-ray scattering beamline to probe coupled spin/charge/orbital excitations in complex materials, and we will continue to develop our world-leading capabilities in ambient-pressure x-ray photoelectron spectroscopy to probe solid/liquid interfaces and to probe heterogeneity in these systems with a nanofocused beam.

The ALS works closely with its users to stay abreast of emerging science areas and to expand the breadth and depth of our experimental capabilities. For example, ALS staff members collaborate actively with scientists in the Lawrence Berkeley National Laboratory (LBNL) Batteries for Advanced Transportation Technologies (BATT) program, the Joint Center for Artificial Photosynthesis (JCAP), and the Joint Center for Energy Storage Research (JCESR) to develop tools to study the electrode/electrolyte interface. The ALS is now part of the LBNL Energy Sciences Area, the heart of DOE BES activity at the laboratory, and regular Area Meetings with partner divisions provide valuable collaborative strategic planning. For example, we work closely with the Chemical Dynamics program in the LBNL Chemical Sciences Division and with the extreme ultraviolet (EUV) lithography program managed by the LBNL Center for X-Ray Optics (CXRO) and supported by SEMATECH. Finally, we collaborate with the Berkeley Center for Structural Biology (BCSB) in the LBNL Physical Biosciences Division to maintain, operate and develop protein crystallography capabilities. These programs are embedded in LBNL divisions but also attract users from around the world to the ALS. The ALS also collaborates closely with several Energy Frontier Research Centers (EFRCs) and a few dozen Approved Programs (APs) from across the country in areas ranging from high-temperature superconductivity to energy and environmental science. In addition to receiving extended access to ALS beamlines, an AP collaborates with ALS staff to develop new capabilities that are made available to the broader user community. This is a valuable mechanism on which we rely very heavily to foster facility development and renewal.

The ALS seeks regular advice on these diverse collaborative programs through several interrelated activities that feed this Strategic Plan. We engage our Users' Executive Committee (UEC) several times per year to discuss how we might help users be more productive at the facility. The UEC also organizes our annual User Meeting, which generally includes 12–15 topical workshops organized collaboratively by our staff and users. These workshops provide invaluable advice on emerging opportunities and research priorities. With oversight from our Scientific Advisory Committee (SAC), the ALS organizes typically two reviews per year of entire subdisciplines to seek focused advice on how to optimize our capabilities to address important research problems. The SAC, composed of national and international experts from many different disciplines, meets twice per year to provide high-level advice on our program.

In addition to motivating the suite of emerging beamline and endstation projects that are the primary focus of this strategic plan, the advisory process described above has helped guide the ALS on a strong record of accelerator improvements and upgrades that have kept the facility at the forefront of soft x-ray science and technology. Implementing top-off injection and a recent sextupole upgrade, for example, increased the source brightness by about a factor of 10 over the past 5 years. An innovation recently made available to users is "pseudo-single-bunch mode," which will expand ALS capabilities in dynamics and time-of-flight experiments. The ALS Accelerator Physics Group has actively participated in the worldwide effort to design ultrahigh-brightness, diffraction-limited storage rings. A major long-range plan is to upgrade the ALS to a multibend achromat lattice that will provide another 100-fold increase in brightness in the soft x-ray regime. This upgrade is directly connected to the scientific focus of this strategic plan since a material's heterogeneity can be encoded into the smooth wave fronts of a diffraction-limited beam; that is, high brightness is directly correlated with our ability to probe heterogeneous materials and devices. The planned upgrade is absolutely crucial to maintaining ALS world leadership in soft x-ray science for decades to come.

The excellent ALS staff has established and maintains a productive and highly collaborative environment to accomplish the ALS mission statement: ***to support users in doing outstanding science in a safe environment.*** Supported by that spirit, this strategic plan outlines a path to a future that is even brighter than our outstanding past.

I. Introduction

The functioning of material devices and biological systems alike often relies on multiscale structures. Defects in an oxide gate a few atoms thick determine how well a transistor functions, which in turn helps determine the speed and power requirements of the gate and thus how many gates can be placed on a chip. Proteins a few nanometers in scale that assemble to form the submicron-scale nuclear pore complex vary their configuration to regulate transport through the nuclear membrane, thereby helping to control cell function. But a multiscale structure by itself does not necessarily do anything useful: a complex correlation between different scales is the key ingredient that endows multiscale structures with useful function. For example, even chemical reactions could not occur but for the complex interplay of high-energy electronic and low-energy vibrational degrees of freedom. These ideas are closely connected to the DOE initiative in mesoscale science: mesoscale structures are functional multiscale systems important in many disciplines.

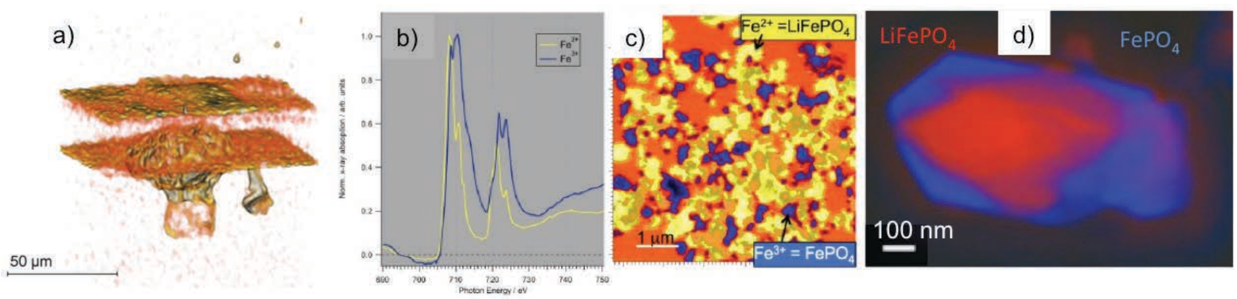

Fig. 1. Spatial scales in lithium ion batteries. (a) Micron-scale x-ray tomography of dendrites forming in a model lithium anode [K.J. Harry et al., *Nat. Mater.* **13**, 69 (2013)]. (b,c) Oxidation-state map of a partially lithiated Li_xFePO_4 cathode measured with a scanning transmission x-ray microscope with ~30-nm resolution [W.C. Chueh et al., *Nano Lett.* **13**, 886 (2013)]. (d) Soft x-ray ptychographic reconstruction of a single grain of a Li_xFePO_4 cathode with < 10-nm resolution (Shapiro et al., submitted).

The ALS vision is to engage our user community to develop and apply cutting-edge tools to probe and control multiscale spatial and temporal correlations at the heart of chemical, biological, and material systems. These tools need to produce multimodal and multidimensional maps to track and understand how chemical or magnetic information is transferred between scales (Fig. 1). The ALS now offers a suite of tools that combine structural, chemical, and magnetic contrast with the ability to map objects over exceptional size and time scales and in diverse environments: small-molecule dynamics, operating electrochemical cells, cycling of batteries and fuel cells, soil samples in various states of hydration, structure and motion of protein molecules in crystals and in solution, subcellular structures, cells, tissues and living organisms, complex electronic and magnetic material devices, and many more. These tools are under continuous development at the ALS and provide crucial capabilities to address DOE initiatives in energy science, mesoscale science, bioscience, and beyond.

Since it was first commissioned in 1993, the ALS has emerged as the world leader in soft x-ray science while also offering complementary hard x-ray capabilities. The facility owes its success to its deep connection to current research needs and trends, driven by an outstanding user population; strong partnership with other LBNL divisions, UC Berkeley faculty, and beyond; continued invention and innovation in instrumentation; increases in source stability and brightness thanks to a successful program of accelerator upgrades; a strong commitment to user support and collaboration; and unswerving attention to all aspects of safety. The facility supports the research of over 2200 users per year whose ALS-based results appear in more than 800 refereed journal publications annually, with over 150 of these articles in high-impact journals. The ALS has 40 beamlines serving users more than 5000 hours each year.

A. ALS Users Address High-Impact DOE Research Problems

Though the ALS is nearly built out, its innovative culture will continue unabated well into the future by continually optimizing existing capabilities, by seeking efficiencies wherever possible, and by repurposing existing beamlines to serve the evolving science needs of our diverse user community. Section II of this strategic plan explains how the ALS will marshal its resources to support aggressive yet cost-effective beamline and endstation projects to serve and further nurture our innovative culture in five key science areas that crosscut the BES Grand Challenges:

- *Mapping electronic, ionic, and chemical pathways in catalysis, energy conversion, and energy storage*: utilize the chemical contrast and spatial resolution of soft x-ray spectroscopy and microscopy to help perfect emerging catalysts and energy materials.
- *Enabling technologies for ultralow-power electronics from EUV technology through emerging quantum, magnetic, and spintronic devices*: utilize the spatial sensitivity and spectral contrast of soft x-rays to help develop new generations of ultralow-power electronic devices in silicon and in multifunctional materials.
- *Illuminating the crossover between dynamic and kinetic temporal regimes at the nanoscale to achieve control over materials synthesis, self-assembly, and function*: understand how fundamental processes like bond breaking and spin flipping connect to activated chemical kinetics and domain-wall motion.
- *Understanding complex interactions and mechanisms across large temporal and spatial scales to understand biological and environmental systems*: harness the power of existing and emerging ALS imaging and spectroscopic tools to understand complex biological and environmental systems, from cells to organisms.
- *Developing mesoscale analog processors from catalytic networks to neural processors*: use the power of soft x-ray spectromicroscopy to help enable an emerging materials area that seeks to build mesoscale devices and machines with targeted function.

These research areas, as well as the strategic plan for beamlines, endstations, and accelerator upgrades discussed below, have been developed and prioritized through extensive engagement with the ALS user community through workshops, cross-cutting reviews, and advisory committees, as explained in Appendix E. These areas evolve synergistically with DOE priorities.

Table 1. ALS Beamline and Major Endstation Construction Projects, 2014–18

Beamline	Project	Commission	Partners & Funding	Sec. II Thrusts Impacted	Notes
7.0.2	Chicane sector, install MAESTRO	2014	SUFD/SISGR, ALS ops, DMSE*	B, E	ARPES/nanoARPES; beamline and endstations with extensive growth capability; mesoscale electronic structures.
4.0.2	5-tesla superconducting octupole endstation	2014	ARRA	B	Magnetic spectroscopy at high field with variable field orientation.
10.0.1.2	Repurpose 10.0.1.2; install advanced spinARPES	2014	ALS ops	B, E	Endstation with enhanced exchange-scattering detectors; enables high-resolution spin-resolved ARPES.
12.0.1, 11.3.2	EUV program upgrades	2013/4	SEMATECH, LBNL	B	New actinic mask inspection station; new high-NA exposure tool; small fab for wafer processing and resist testing.
2.4	Infrared spectrotomography	2014	SUFD/NSLS, BER/BSISB	D	Increase IR capacity; infrared beamline for spectromicroscopy of environmental and biological samples; IR tomography.
7.0.1	COSMIC	2015	DMSE, ALS ops, MF/NCEM	A, B, C, E	Coherent scattering and imaging beamline; mesoscale chemical imaging; XPCS in complex magnetic systems.
2.0	Clear sector, install GEMINI	2016	HHMI, LBNL, LBNL/PBD	D	High-brightness cryogenic undulator for small crystals and large unit cells; advanced detectors; robotic sample handling.
6.0.2	Repurpose 6.0.2; install AMBER	2016	PNNL, JCAP, BATT, JCESR, ALS ops	A, E	Energy materials beamline: soft x-ray absorption and emission, STXM, APXPS with diverse and flexible sample environments.
6.0.1	Repurpose 6.0.1; install QERLIN	2017	ALS ops	B, C, E	Soft x-ray resonant inelastic scattering beamline to probe coupled excitations in complex materials.
11.3.1	Move chemical crystallog. to SB; install sample robot	2016	LBNL/MSD, LBNL/CSD, ALS ops	A	100x higher flux at high energy; diverse sample environments. Install robot for efficient material screening (seeking partner funding).
9.0.1/2	Move SXR branch; chicane in future	2016	CSGB, LBNL/CSD	A, C	Doubles capacity; expands capability into soft x-rays; allows flexible studies of ultrafast dynamics (seeking to partner with LBNL/CSD/CSBG for funding).
9.3.1	Upgrade aging tender-energy beamline	2016	CSGB, LBNL/CSD	A	Doubles capacity; expands capability into soft x-rays; allows flexible studies of ultrafast dynamics (seeking partner funding from JCAP, JCESR, BATT).
7.3.3	Move materials SAXS to wiggler	2017	ALS ops	A	100x increase in flux and capacity; increase time resolution and install crystal monochromator for lower-energy bandwidth; diverse sample environments.
4.0.2	Magnetic spectroscopy beamline upgrade	2017	ALS ops	B	15-year-old beamline, 10x more flux and brightness with optics upgrade and optimized undulator period.
?	Tender-energy spectroscopy and scattering	2018	LBNL/ESD, SIBYLS, BCSB	D	New soft/intermediate energy SAXS capability to probe biological and environmental structures in diverse environments .
6.1.2, 6.3.1	Magnetic microscope	2018	ALS ops/ LDRD	B	Replace or upgrade XM-1; spectromicroscopy; variable temperature and magnetic field.

Commissioning — Funded / in procurement — Mostly funded / in design — Future planning / not funded

* List of acronyms can be found in Appendix A.

B. Strategic Beamline and Endstation Projects Address the DOE Mission and ALS Priorities

Table 1 summarizes strategic beamline and endstation projects having a total cost either known or estimated to be in excess of $1M. The table delineates projects that are being commissioned, are funded and under construction, are partly or fully funded and are being designed, and those that are our highest-priority beamline projects and are planned over a longer time scale. It also indicates the diversity of partners that are helping to develop the ALS program by contributing financially to these projects (a list of acronyms can be found in Appendix A).

The fifth column of Table 1 is key: it connects the various beamline construction projects to the research areas in the subsections of Sec. II. Each subsection explains the importance of a particular research area, indicates how soft x-ray science and technology will play a major role in addressing key underlying issues, provides a brief description of existing ALS capabilities relevant to the area, and discusses how near- and long-term beamline and endstation strategic plans provide crucial future capabilities.

While the ALS continues to innovate and upgrade its capabilities, the facility carefully balances its suite of instruments with the staff it is able to support so as to maintain efficient and sustainable operations. The ALS maintains a spreadsheet of beamline metrics that provides a snapshot of beamline staffing, user demand, usage, operational complexity, and overall productivity. This is used to help maintain an appropriate level of support on different beamlines. We increasingly train our staff across different beamlines to enhance our operational efficiency. Most of the projects listed in Table 1 are net-staffing-neutral.

Table 2. ALS Accelerator Construction Projects, 2014–18

Project	Commission	Notes
Storage ring rf upgrade	2014	Update aging rf system; eliminate potential major single-point failures (nearly complete).
Storage ring controls upgrade	2014	Update aging storage ring control system (nearly complete).
Pseudo-single-bunch operation	2014	Bumps a single electron bunch out of the normal orbit using “kick and cancel” approach; enables timing and dynamics studies; ready for user ops in 2014.
AC power conditioning	2018	Addresses external power glitches, which are the source of most ALS beam losses.
HVAC/energy efficiency	2017	Stabilizes ALS environment for precision optics and endstations.
Undulator technology	ongoing	Work on advanced undulator designs for the future.

C. Strategic Accelerator Upgrades Enable Continuous Improvement in ALS Tools

Cutting-edge endstations and beamlines are only part of what is required for continuous ALS renewal and thus do not capture the totality of the ALS innovative spirit. Section III.A describes an ongoing, aggressive process of accelerator renewal and upgrade. The ALS is emerging from a phase of renewal that modernized most major existing accelerator systems: power supplies, control systems, rf system, and a host of smaller components. Conversion to top-off operation and the addition of 48 sextupoles to the lattice has increased source brightness by an order of magnitude in the past six years. Remaining strategic priorities for the existing ALS accelerator for 2014–18 are summarized in Table 2 and elaborated in Sec. III.A and Sec. III.B.

Finally, a crucial long-term focus of our strategic plan, which will ensure that the ALS continues world-leadership in soft x-ray science and technology for decades to come, is to replace the ALS accelerator with a multibend achromat lattice that will provide ultrabright, diffraction-limited soft x-ray beams up to ~2 keV. This planned upgrade, elaborated briefly in Sec. III.C, is in perfect sync with this strategic plan since there is a direct relationship between source transverse coherent power and our ability to probe multiscale heterogeneous systems.

Table 3. ALS Ancillary Support Projects, 2014–18

Project	Commission	Notes
User portal	2014	Complete modern portal to ensure efficient and safe user processing and service.
LUXOR	ongoing	Ongoing optics upgrade for older ALS beamlines.
Metrology lab	2014	New/upgraded instruments to complete Optics Metrology Lab.
Scientific data portal	ongoing	Maintain and expand our ability to transmit, store, manage, and analyze large data sets.
Detector projects	ongoing	Continue development of detectors with high frame rate, high spatial resolution, or other specific characteristics.
X-ray streak cameras	ongoing	Enhance existing x-ray streak camera program to serve ALS nanokinetic programs.
Fellowship programs	ongoing	Ongoing support of ALS post-baccalaureate, doctoral, and postdoctoral fellowship programs for professional and staff development.

D. Ancillary Capabilities to Support a Strong Science Program

The appendices of this document describe several enabling ALS programs and technologies that support our strategic research priorities. These include maintaining a strong safety culture, focusing on ALS staff professional development, engaging users in the strategic planning process, testing and regularly upgrading beamlines and optics, developing and deploying state-of-the-art x-ray detectors, building the infrastructure needed to manage and analyze the huge volume of data produced at the ALS, and establishing efficient procedures to optimize facility usage and facilitate a smooth user experience. Ongoing projects in these activities, which are summarized in Table 3, require real resources and are therefore managed strategically and balanced against other instrument and accelerator needs.

II. ALS: Ideal Tools to Probe Functional Materials and Devices

A. Mapping Chemical and Energy Pathways

Devices currently in use or being developed for selective and efficient heterogeneous catalysis, photocatalysis, energy conversion, and energy storage rely heavily on diverse multiscale phenomena, ranging from interfacial electron transfer and ion transport, occurring on nanometer to picosecond scales, to macroscale batteries that charge in hours and catalytic reactors with turnover rates of ~1/s. Soft and hard x-rays can probe dense environments with atomic and chemical contrast spanning a large spatiotemporal range, thereby providing unique fundamental information about functioning mesoscale devices. For example, in the next year ALS staff will collaborate with user groups to develop sample environments so the battery oxidation-state map in Fig. 1(c) can be accomplished in real time during charging cycles. Such "nanokinetic" measurements are essential to optimizing such complex multiscale (electro)chemical devices.

The number of current ALS beamlines used by users involved in energy science and catalysis is extensive, ranging from a productive SAXS/WAXS beamline through our cutting-edge scanning soft x-ray transmission microscopes (STXMs). Many of these are among our most heavily over-subscribed beamlines and Table 1 indicates that our strategic priorities increase both our capability and our capacity to help users study energy and catalytic materials.

Cutting-edge spectromicroscopy: COSMIC. Within two to three years ALS users will be able to probe the large-scale structure of battery dendrites [Fig. 1(a)] with x-ray tomography and zoom in with COSMIC to do a chemical map to understand failure mechanisms [Fig. 1(b,c)] or probe the interface region around an individual electrode grain [Fig. 1(d)]. Since 1995, the ALS has led the world in developing STXMs, and the STXMs on ALS Beamlines 5.3.2.1 and 11.0.2 are highly productive [e.g., Fig. 1(b, c)]. Commissioning of the Coherent Scattering and Micrscopy (COSMIC) beamline in early 2015 will maintain this world leadership. One branch of COSMIC will do ptychographic diffractive imaging with state-of-the-art scanning systems, high-data-rate CCD detectors matched to a high-bandwidth data system, and diverse in situ sample environments. The precursor of COSMIC provided the image in Fig. 1(d). COSMIC will provide images with a resolution of a few nanometers, ultimately in 3D with full chemical contrast. Among many other areas, this will revolutionize our ability to probe "interphase" regions like the solid-electrolyte interphase that governs the operation of batteries, fuel cells, and other electrochemical systems as well as photocatalytic reactors used for artificial photosynthesis.

In addition to the advanced imaging capabilities exemplified by COSMIC, ALS development of in operando sample-handling systems, high-throughput x-ray emission spectrographs, and ambient-pressure x-ray photoelectron spectroscopy (APXPS) has attracted many users wanting to tackle key basic-science issues in catalysis and energy sciences. Soft x-ray absorption (XAS) and emission (SXE) spectroscopies on Beamline 8.0.1 provide a direct probe of a material's

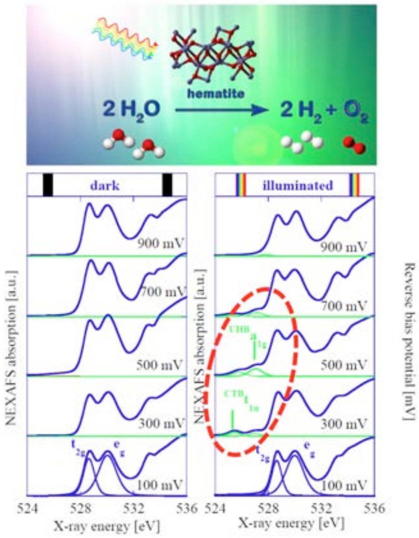

Fig. 2. Electron–hole formation at the hematite/water interface upon absorption of light. A. Braun et al., *J. Phys. Chem C* **116**, 16870 (2012).

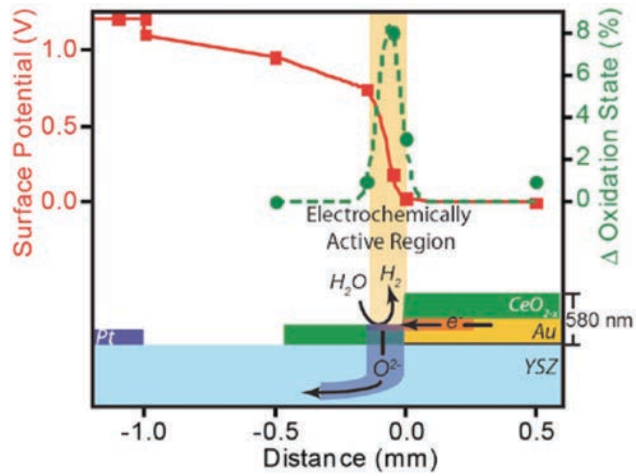

Fig. 3. Mapping the local potential in a solid-oxide fuel cell using core-level shifts with APPES (*Nature Materials* **9**, 944 (2010).

electronic levels that are directly involved in chemical bonding, in diverse environments and often in functioning devices. For example, the JCAP Energy Hub uses them heavily to understand the fundamental interfacial processes in candidate artificial photosynthetic cells (Fig. 2). APXPS has been developed on Beamlines 9.3.1, 9.3.2, and 11.0.2 to probe, for example, catalytic reactions under realistic operating conditions and electrode surfaces in contact with an electrolyte and is now being deployed at facilities around the world. An important recent result (Fig. 3) demonstrated the ability to map directly the local electrostatic potential in an operating solid-oxide fuel cell.

Multimodal chemical analysis: AMBER. A strategic priority is to collect these spectroscopies and microscopies on a single beamline for multimodal analysis of energy and catalytic systems. Toward this end, we will repurpose Beamline 6.0.2 to install the Advanced Materials Beamline for Energy Research (AMBER). Beamline 6.0.2 is presently one of two branches of the femtosecond laser-slicing program at the ALS. The ALS and LBNL are proud to have played a major role in developing ultrafast x-ray science, but much of the excitement in that area has migrated to FEL sources, and it is time to redirect our resources to serve larger communities more closely aligned with ALS capabilities. AMBER will provide in situ sample preparation with SXE/resonant inelastic x-ray scattering (RIXS) and absorption spectroscopies, APXPS with high spatial resolution at near-atmospheric pressure, and a high-throughput STXM capability. Construction of AMBER will be partially supported by the JCAP and JCESR Energy Hubs, the LBNL BATT program, and by partners at PNNL, all of which are highly motivated by the assembly of AMBER tools to study and chemically map materials in diverse environments.

Other planned beamline upgrades. The ALS also operates a unique resonant soft x-ray scattering (RSoXS) beamline (11.0.2) applied mostly to soft materials, a complementary capability to the workhorse hard x-ray SAXS/WAXS beamline (7.3.3), and a materials crystallography beamline (11.3.1). These are heavily used to study energy materials in unusual environments, for example, microporous membranes used in batteries, fuel cells, artificial photosynthetic devices and metal-organic framework (MOF) compounds proposed for gas separations, carbon sequestration, and catalytic reactors. Planned upgrades include the following:

> **RSoXS upgrade to probe liquid environments:** The ALS RSoXS beamline was commissioned three years ago and has used the chemical contrast available near the carbon K-edge to probe organic and polymer thin films, notably the interfacial structure of organic transistors and heterojunction photovoltaics. An upgrade to study liquid and complex fluid samples in several different environments is planned for this year.
>
> **SAXS/WAXS upgrade:** In the next three to four years, we plan to relocate the SAXS/WAXS Beamline 7.3.3 to a superbend or possibly the 5.0 wiggler source with 100x higher flux at high energy. This will allow either sub-ms time-resolved studies or higher-energy-resolution studies of bulk materials and thin films. The endstation supports an increasingly diverse set of sample environments and is a pilot project in our effort to handle large data streams.
>
> **High-throughput materials crystallography:** We are seeking partners in the LBNL Materials Sciences and Chemical Sciences Divisions to help fund migration of Beamline 11.3.1 to a superbend over the next two to three years. This will provide ~1000x more flux at high energy to increase capacity, precision, and other capabilities. In the next one to two years we will install robotic sample handling to enhance throughput and allow rapid materials screening and combinatoric experiments. This very productive beamline and the SAXS/WAXS beamline are increasingly used in “mail in” and/or “remote access” modes.

B. Enabling Ultralow-Power Electronics through Multifunctional Materials Development

The ALS supports diverse activities along the path between the fundamental electronic and magnetic properties of materials to functional electronic and magnetic devices that are very close to market. Understanding the fascinating materials physics of multiferroics, oxide superconductors, graphene, topological insulators, and skyrmions, for example, is often related to an important practical goal: to reduce the power consumed by electronic devices ranging from transformers to microprocessors. Most future applications will involve multiscale devices that channel a signal from a nanoscale structure—a few electrons on a capacitor or a nm-scale magnetic bit, for example—into the macroscopic world with high efficiency, high fidelity, and low noise. Solving this electronic power consumption problem will require development of new transformative technologies that enable ultralow-power logic elements, memory devices, power conversion devices, and beyond. ALS users with interests in electronic devices and quantum and magnetic materials use the ALS in research that supports or fosters many of the candidate technologies.

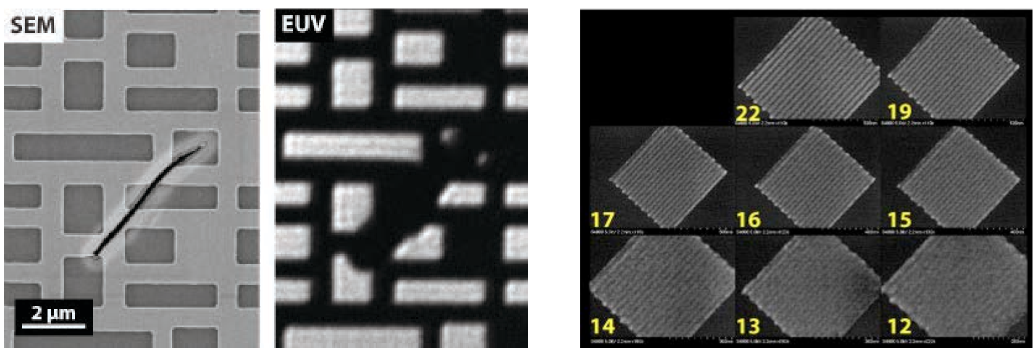

Fig. 4. Left: Imaging mask defects with secondary electron microscopy and an EUV actinic inspection tool. At-wavelength imaging is crucial to characterize amplitude and phase defects in multilayer masks used in EUV lithography. [Mochi, et. al., *Proc. SPIE* **7636**, 76361A (2010)]. Right: Lines and spaces printed with EUV lithography down to a 12-nm period. Such studies have been essential in developing EUV optics and resist materials.

Emerging nanoscale circuits: EUV lithography. The microelectronics industry has managed the power problem primarily by making smaller, lower-power transistors. Worldwide photolithography research hinges on an abrupt jump to a radically shorter wavelength range known as extreme ultraviolet (EUV), with the promise of nanoscale circuit patterns and generations of continued shrinking. Synchrotrons are among the world’s brightest sources of EUV light, and the ALS, through the LBNL Center for X-Ray Optics, has emerged as a unique resource for pre-competitive EUV lithography research and technology development, fostering a number of breakthroughs over the past 15 years (Fig. 4). World-leading research programs performed in part at the ALS in optics, masks, photoresist materials, and thin-film mirror-coating technologies have defined the state of the art in this field.

Table 1 indicates that SEMATECH is funding a major expansion of the CXRO EUV program, with installation in 2013 of a new, 100x brighter tool for at-wavelength inspection of EUV reflection masks, and in 2014 a new EUV exposure tool with a resolution limit of 8 nm coupled to a clean room with state-of-the-art semiconductor materials processing equipment.

Smaller microelectronic devices operate with lower power, but this is more than offset by the ever-increasing density of transistors on a chip: the power dissipated by a microprocessor has increased by typically 20% per year even though the energy consumed by a single logic operation has fallen exponentially with Moore's law. For this reason there is a pressing need to develop materials that enable ultralow-power electronic devices. The ALS is having a major impact in this area, and Table 1 indicates that even more powerful tools are planned.

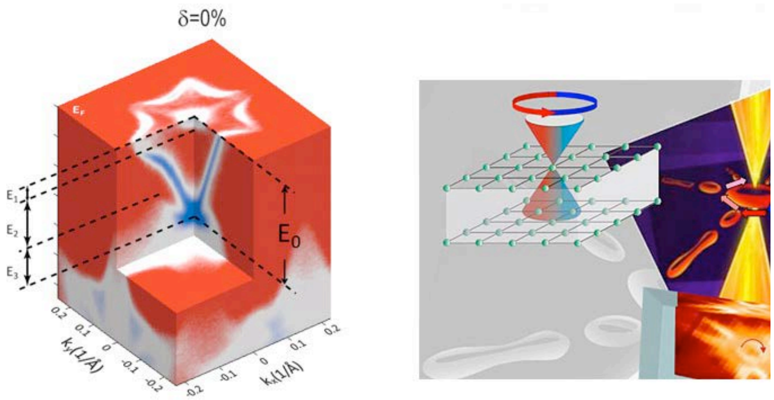

Fig. 5. First demonstration of a topological insulator by high-resolution photoemission. D. Hsieh et al., *Nature* **452**, 970 (2008), and Y.L. Chen et al., *Science* **325**, 178 (2009).

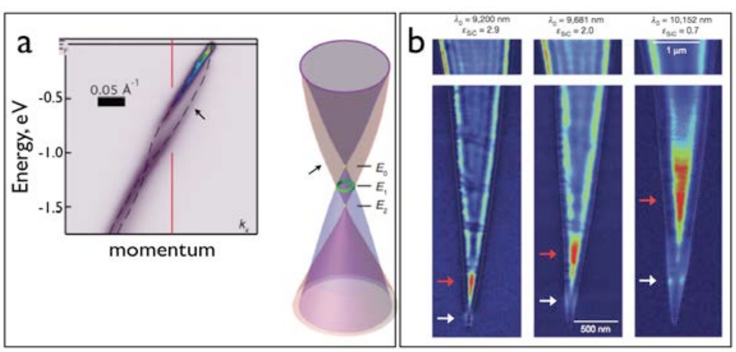

Fig. 6. (a) ARPES spectrum for a homogeneous sample of graphene shows the presence of a satellite band (indicated by the black arrow) due to plasmaronic states. Bostwick et al. *Science* **328**, 999 (2010). (b) Graphene nanostructure designed to confine plasmons (red/white arrows). Chen et al., *Nature* **487**, 77 (2012). With spatial resolution, ARPES can be used to investigate the spatial dependence of electron–plasmon coupling.

Probing electronic structure on the mesoscale: MAESTRO. The angle-resolved photoelectron spectroscopy (ARPES) program at the ALS has led the world to many important discoveries relating to pure and homogeneous materials, including work in high-temperature superconductivity and giant magnetoresistance, as well as the discovery of the properties of graphene, topological insulators (TIs), and other exciting new "Dirac" materials (Figs. 5–6). These discoveries were made possible by a continuous and ongoing program of instrumentation development in detectors, in situ sample preparation, facile data handling and analysis software, and cryogenic sample goniometry.

In the past few years, we have worked to extend this capability to probe the interplay of structure—either externally imposed through material engineering or through self-organization—with electronic properties. This has culminated in the construction of the Microscopic and

Electronic Structure Observatory (MAESTRO) beamline (Table 1), which will improve the spatial resolution available at the ALS by a factor of about 1000, down to 30 nm. Coupling such a probe to extensive thin-film growth and ancillary characterization tools, we will be able to address major problems such as the origin of self-organized structures in correlated materials; the implementation of high-mobility materials such as TIs and metal-oxide heterostructures in novel device schema; the examination of novel electronic materials such as graphene in high-field devices; the probing of multifunctional materials on the nanoscale; electronic structure of nanocrystals on an individual particle basis; and the coupling of light and electronics in emergent “plasmonic” technologies (Fig. 6).

Transition-metal oxides exhibit an expanding array of properties driven by complex coupling between low-energy excitations—phonons, plasmons, spin waves, orbital excitations—and the electron gas. Developing a detailed understanding of these material properties remains a challenge at the forefront of condensed matter physics that will help us understand transport and phase behaviors and thereby enable deployment of ultralow-power electronics in the coming decades. The ALS ARPES program, augmented by the MAESTRO project discussed above and the spinARPES described below, will ensure continuing broad impact well into the future.

Coupled excitations in complex materials: QERLIN A strength of ARPES is that it measures the coupling electrons and holes to low energy excitations, but the results are integrated over all low energy excitations. It can be difficult to verify which excitation(s) lead to a particular exotic property. To understand what drives a particular property – high temperature superconductivity, for example – it is crucial to measure the dispersion relations of the low energy excitations directly, with high resolution, with soft x-ray contrast, and over a large region of Fourier space. For this reason, and also for the connection to nanokinetics discussed in the next section, one of the highest ALS priorities is to develop a soft x-ray RIXS beamline called QERLIN (Q- and Energy-ResoLved INelastic scattering). We will repurpose Beamline 6.0.1, the second part of the ultrafast slicing program, to develop QERLIN. QERLIN will be based on a cutting edge optical design that involves multiplexing the incident beam across the face of the sample, providing 10’s of meV resolution with good signal in a very cost-effective approach.

Phase behaviors and anisotropies in complex materials: High-field magnetic spectroscopy. Resonant soft x-ray spectroscopy and scattering have also emerged as important tools in probing spin-, charge-, and orbital-ordered ground states of transition-metal oxides, and the ALS has important capacity in this area as well. Table 1 includes an important augmentation of that capacity with an ARRA-funded superconducting octupole endstation. This is presently being commissioned and will enable magnetic spectroscopy of oxides and hard magnet phases with fields up to 5 T in arbitrary directions, thereby allowing users to probe the poorly understood anisotropy of spin- and orbital-ordered phases in complex oxides.

There is rapidly growing convergence between the primarily microelectronic technologies described above and emerging magnetic and spintronic technologies. ALS users are playing a major role in illuminating and facilitating that convergence. Magnetism constitutes an enormously important energy and energy-conservation technology as well, being used in transformers, motors and generators, information storage, and magnetic logic elements. Unlike charge currents, spin currents travel in materials without dissipating energy, and furthermore the spins may be used to store information. Spintronics, the manipulation of spin currents in devices, not only promises to consume less energy than today's electronics, but can also enable new technologies like quantum computers and cybersecure encryption schemes.

Magnetic and spintronic materials can be ideally studied with polarization-dependent soft x-ray techniques, since these provide quantitative magnetic information with element specificity, sensitivity to the valence state of the absorber, and the symmetry of the absorber site. Moreover, soft x-ray magnetic microscopies provide nanometer spatial resolution, and time-resolved measurements allow access to fundamental time scales. Over the past 15 years, scientists at the ALS have invented, constructed, and optimized unique spectroscopy, microscopy, and scattering instruments that represent new experimental capabilities and provide access to new experimental geometries that had not been explored before. Table 1 includes several strategic projects that will enhance ALS capabilities in magnetism and spintronics.

Internal spin sources and spin currents: SpinARPES. A key enabling issue in spintronics is to develop internal sources of spin currents. TI states, giant spin Hall structures, and half-metallic compounds are popular candidate technologies that are all actively studied by ALS users. A new spinARPES spectrometer based on ALS-developed high-efficiency spin detectors is presently being commissioned on Beamline 10.0.1. We will also continue to develop time-of-flight spinARPES capabilities used during two-bunch and possibly pseudo-single-bunch operation.

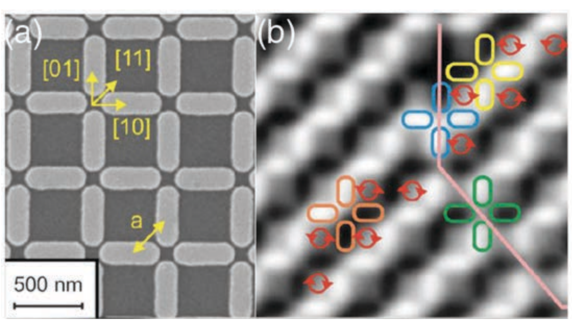

Fig. 7. (a) SEM image of permalloy artificial spin-ice structure. (b) PEEM image resolving defect magnetic configurations (Farhan et al., *Phys. Rev. Lett.* **111**, 057204 (2013).

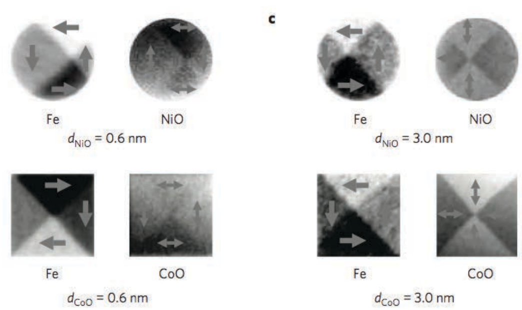

Fig. 8. Ferromagnetic and antiferromagnetic vortex states in NiO/Fe (top) or CoO/Fe nanodiscs (bottom). Wu et al., *Nat. Phys.* **7**, 303 (2011).

X-ray spectromicroscopy of magnetic and spin structures: Magnetic x-ray microscope. An important longer-term strategic priority is to develop a flexible beamline optimized for soft x-ray spectromicroscopy of magnetic materials. Some capacity in this area exists in ALS existing microscopes, but the difficulty of precision control systems with variable temperature and magnetic field and the difficulty of photoelectron emission microscopy (PEEM) with an applied field seriously limits our capacity to study magnetic materials in situ and magnetic devices in operando. Examples of systems that can be only partly studied include magnetic domain behaviors, spin ice and other patterned nanomagnetic structures (Fig. 7), ferromagnetic and antiferromagnetic vortices (Fig. 8), and skyrmions. ALS user groups are actively studying all of these kinds of systems using magnetic spectroscopy and scattering, x-ray microscopy, and PEEM. The COSMIC scattering branch, in Table 1 and described in more detail in the following section, will provide a valuable probe of equilibrium or steady-state magnetization dynamics. But the limit in ALS capability is serious; for example, most known skyrmion phases exist below room temperature and at moderate applied field, and these cannot presently be studied on existing ALS microscopes. The ALS is considering including in its portfolio a soft x-ray spectromicroscopy beamline dedicated to magnetic and spintronic materials (Table 1) with about 15- to 20-nm spatial resolution and the ability to vary the temperature and applied magnetic field. This might entail an upgrade of the full-field microscope on Beamline 6.1.2, or it could be a STXM on Beamline 6.3.1 or 4.0.2.

Multifunctional materials discovery: Combinatoric endstation. An increasingly popular and important approach to discovering new materials is to produce combinatoric libraries, either discrete samples or composition gradients. Such a program is particularly important in magnetic materials given their importance in many energy conversion technologies, but clearly numerous long-range goals also exist in functional oxides, lanthanide compounds, and beyond. A key feature of these efforts is to apply cutting-edge microfocused tools to probe the desired material properties quickly and efficiently, coupled to strong theory and modeling programs. For magnetic and spintronic materials, the first of these is presently being developed on ALS magnetic spectroscopy Beamline 6.3.1, which is also being upgraded to provide a smaller focal spot and equipped with the necessary sample handling and, in the future, in situ growth. Also, a connection is being formed with the Materials Project at LBNL (see *https://materialsproject.org/*) to provide valuable computational and theoretical guidance for this program. This effort has been planned for some time, but the rapid implementation was strongly supported by a recent crosscutting magnetism review.

C. Illuminating the Crossover between Atomic-Scale Dynamics and Nanoscale Kinetics

A hierarchy of length and time scales governs many important dynamical processes. For example, reconfiguration of small molecules often occurs on picosecond vibrational or femtosecond electronic time scales. In a protein molecule, however, ultrafast dynamics near a reaction center can be dramatically influenced by the longer-scale conformational changes of the protein backbone. Similarly, a spin flip at a magnetic domain wall, for example, can occur on the picosecond time scale characteristic of spin waves, but the domain wall moves much more slowly and the domain wall configuration helps control the flipping of a single localized spin.

In chemical reactions, the dynamic–kinetic crossover occurs roughly at a time scale of $h/k_BT \sim 1$ ps. A process occurring much faster than this is dynamical, and temperature and Arrhenius kinetics are not relevant. A slower process is inherently kinetic and is often modeled statistically with, for example, a prefactor and activation energy. By contrast, the dynamical oscillatory modes of a magnetic nanostructure can persist for times beyond 1 ns, and diffusive motion characterized by kinetic rate equations dominates at longer time scales. We call the regime between these dynamical and kinetic limits the "nanokinetic" regime, which is the focus of this third ALS research theme. We seek to develop soft x-ray tools to probe nanokinetics, to establish rules that govern the interplay between dynamical and kinetic phenomena and to gain control over how a system evolves through the nanokinetic regime.

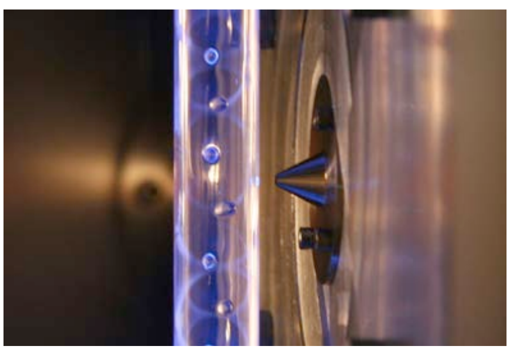

Fig. 9. The chemical reactions of Criegee intermediates important for atmospheric chemistry were studied using an apparatus at ALS Beamline 9.0.2. O. Welz et al., *Science* **335**, 224 (2012).

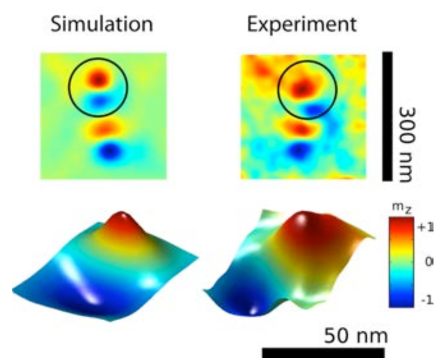

Fig. 10. Stroboscopic images of the switching of coupled magnetization vortex cores in permalloy discs, measured on the ALS Beamline 11 STXM. A. Vansteenkiste et. al, *Nat. Phys.* **5**, 332 (2009).

The ALS has an array of tools to undertake such studies. The productive and unique ALS Chemical Dynamics program, for example, combines ALS capabilities with resources and expertise in the LBNL Chemical Sciences Division to serve an important and large user community. Examples of recent accomplishments include identification of the Criegee intermediate in combustion with spectroscopy in a flame (Fig. 9) and identifying a new path for hydrogen bonding and proton migration. In a very different context, a decade ago ALS staff and users invented tools to probe magnetization dynamics with time-resolved STXM, PEEM, and full field x-ray microscopy. This strong ALS community has focused recently, for example, on magnetization dynamics in single and coupled magnetic nanostructures (Fig. 10).

A key aspect of the current ALS strategic plan is the termination of our existing programs in Sector 6 based on ultrafast x-ray pulses produced with laser slicing of the electron beam. Activity in this area is rapidly migrating to FEL facilities, and ALS resources will be repurposed to serve communities more closely aligned with the capabilities of ring-based sources. This decision is accompanied by a plan to renew and expand our focus on nanokinetic phenomena. Several of the strategic capabilities in Table 1 reflect this change of focus and will focus directly on nanokinetic problems.

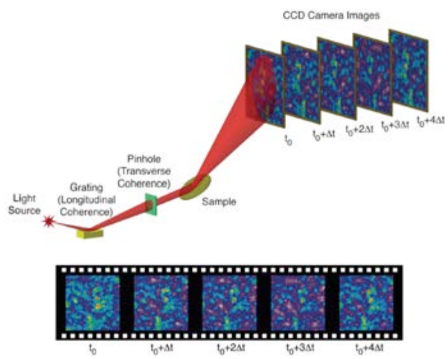

Fig. 11. Thermally driven fluctuations in the orbital-ordered phase of a complex manganite crystal using coherent soft x-ray scattering. J.J. Turner et al., *New J. Phys*. **10**, 053023 (2008).

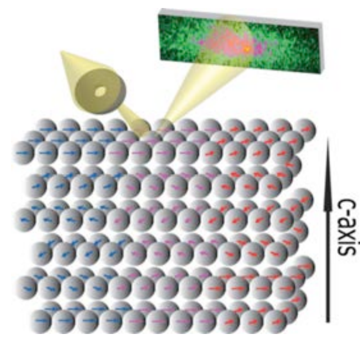

Fig. 12. Equilibrium magnetization fluctuations and domain jamming was studied in antiferromagnetic dysprosium on ALS Beamline 12.0.2.2 using XPCS. S.W. Chen et.al., *Phys. Rev. Lett*. **110**, 217201 (2013).

Nanokinetics with COSMIC. In addition to the ptychography capability discussed in Sec II.A, COSMIC will also provide a branch for soft x-ray photon correlation spectroscopy (XPCS, Fig. 11). This will be ideal for measuring nanoscale kinetic phenomena with resonant soft x-ray contrast to probe equilibrium, steady-state, and nonequilibrium processes in chemical and magnetic systems. Experiments on the precursor beamline to COSMIC (Beamline 12.0.2.2) have focused primarily on complex magnetic systems (Fig. 12), but we plan to expand to chemical systems to measure, for example, catalytic kinetics in nanoporous materials like zeolytes, thereby connecting the science goals in this section to those in Sec. II.A. A crucial goal is to increase the spatiotemporal dynamic range of the technique, which depends quadratically on source brightness and coherent flux. COSMIC will expand our capabilities, but the lattice upgrade described in Sec. III.B will provide a revolutionary expansion of XPCS.

Connecting XPCS/COSMIC with RIXS/QERLIN to probe nanokinetics. Van Hove’s space-time correlation formalism of neutron and photon scattering indicates a direct relationship between correlation spectroscopy in the time domain and quasi-elastic scattering in the frequency domain. Indeed, the shape of the quasi-elastic neutron scattering peak is commonly used to probe diffusive fluctuations of overdamped material modes. In the same way, RIXS can used to probe subpicosecond kinetics in Fourier space. A key long-range goal of the ALS strategic plan is to connect the sensitivities of the QERLIN and COSMIC scattering beamlines to provide a revolutionary tool set to probe the nanokinetic regime.

Supporting technologies to probe nanokinetics. The first x-ray streak camera was commissioned at the ALS about 10 years ago and remains a useful capability. The change of ALS focus from ultrafast x-ray science to nanokinetics suggests numerous applications for high-frame-rate streak cameras with ~1 ps time resolution. For this reason we plan to re-energize our streak camera development program to focus on both externally stimulated and thermally driven processes. This effort will need to be closely coupled to efforts focused on developing fast-readout detectors—MHz and faster—since this will often determine the repetition rate that can be used. Streak cameras also offer interesting prospects to fast XPCS measurements.

Accompanying ongoing detector development will be the development of new ways to pump a sample. The ALS has already collaborated with users from the LBNL Chemical Sciences Division to purchase a fast laser that is synced to the storage ring pulses and that is available to users around the ring. Beyond that we have in the past developed fast switches and strip-lines to provide fast magnetic-field pulses. We will expand the availability of this and other fast-pump technologies to allow users to apply diverse soft x-ray spectroscopies and microscopies to pobe the nanokinetic regime.

D. Untangling Complex Interactions in Biological and Environmental Systems

The ALS supports tools to image biological structures ranging from biopolymer molecules using macromolecular crystallography to entire organisms using x-ray tomography (Fig. 13). A similar range of environmental structures can also be probed with chemical contrast. As in the other research areas discussed here, biological and environmental systems exhibit interesting and important phenomena that involve interaction between scales. The activity of an enzymatic center is determined by the secondary and tertiary structures of a protein molecule. The complex chemistry of a soil particle is determined by the availability of various species at the interfaces.

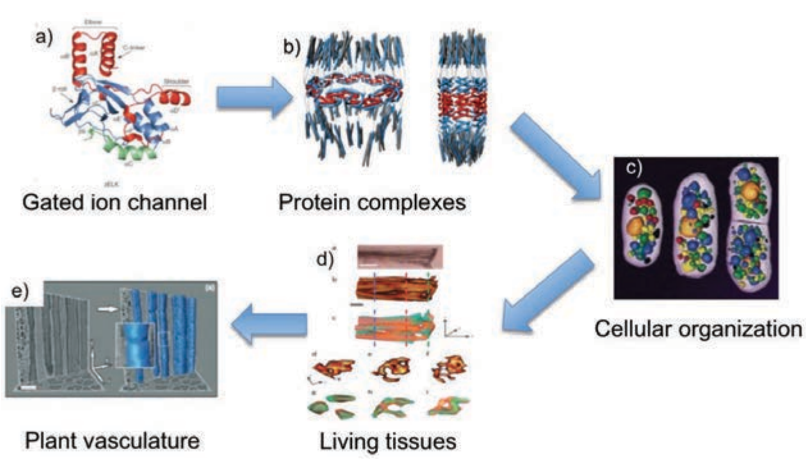

Fig. 13. The scales of structural biology illuminated by recent ALS results: (a) protein structure of a gated ion channel [Brelidze et al., *Nature* **481**, 530 (2012)]; (b) protein assembly to form the nuclear pore complex [Solmaz et al., *PNAS* **110**, 5858 (2013)]; (c) single-cell CAT scans with soft x-ray nanotomography [Parkinson et al., *J. Struct. Biol.***162**, 380 (2008)]; (d) infrared tomography of living tissues with few-micron resolution [Martin et al., *Nat. Meth.* **10**, 861 (2013)]; (e) x-ray tomography of living grape vines [McElrone et. al., *J. Vis. Exp.* **74**, e50162 (2013)].

The capabilities of soft and hard x-ray techniques to address biological problems pertain mostly to the images in Fig. 13, not the arrows that connect different scales; yet these transitions between scales are of crucial importance to understanding how these complex systems function. What determines the function of a given protein molecule in a complex? How is a protein complex distributed in a cell to support and regulate cell function? How does intracellular signaling make a tissue work? We have many tools to measure biological and environmental structures over a large spatial scale, but we have mostly an empirical understanding of how these scales interact. Addressing these questions will illuminate the connection between structural and systems biology, or environmental structures and environmental systems. Emerging tools for environmental and biological science in Table 1 will help fill this crucial gap in understanding.

GEMINI. The macromolecular crystallography beamlines at the ALS have enabled outstanding scientific productivity, providing high-performance hard x-ray diffraction beamlines that have kept pace with the changing needs of the structural biology community [e.g., Fig. 13(a)]. To continue to provide the highest possible performance, the Howard Hughes Medical Institute has recently funded a new high-brightness protein crystallography facility called GEMINI at ALS Sector 2. This will significantly expand our ability to probe small crystals with large unit cells, with an emphasis on protein complexes like the nuclear pore complex in Fig. 13(b). GEMINI will include a high-brightness cryogenic undulator serving two branches simultaneously located in a single hutch. One of the branches will be served with diamond beam-splitters and operate at fixed wavelength; the other will be variable wavelength for multiwavelength anomalous dispersion measurements. Design is underway and completion is expected in three years.

Infrared spectrotomography of living systems. SUFD provided funds to increase ALS infrared capacity to help the NSLS community through the "dark age" between the shutdown of NSLS and the commissioning of IR beamlines on NSLS-II. Using these funds, we will develop source point 2.4, reflecting the beam to the Beamline 1.4 IR endstations so that two stations can be used simultaneously. We have worked closely with NSLS staff to develop this plan and they have submitted an Approved Program proposal to serve their needs and to establish a presence on the ALS floor. The new IR beam will serve a full-field microscope with a focal plane array detector. Moreover, ALS staff collaborated with Carol Hirschmugl of the Synchrotron Radiation Center at the University of Wisconsin to develop infrared spectrotomography [Fig. 13(e)]. This new IR station will be able to do full-field microscopy and tomography, thereby providing a new and valuable way to produce 3D images of living biological and environmental systems with ~5 micron resolution and the chemical contrast of infrared spectroscopy. ALS and NSLS IR communities alike are very enthusiastic about this future; an ALS User Meeting IR workshop in October 2013 overflowed for two full days.

Biological SAXS. Protein SAXS on the ALS SIBYLS beamline, with a focus on combined SAXS/crystallography studies, is highly productive and there is a need for more capacity in this area. There is also interest in building a new soft/intermediate x-ray energy scattering beamline. The higher scattering rate at soft x-ray wavelengths will enable the highest-quality time-resolved measurements from small quantities of samples. Such kinetic measurements help address the structural/systems biology interface discussed above. Grazing-incidence scattering provides significant information on membranes and membrane proteins. Developing these capabilities over the next several years will depend on funding raised by the bioscience community.

Two smaller ALS innovations will also impact biological and environmental sciences:

Nano-infrared imaging. The spatial resolution of far-field infrared imaging and spectro-microscopy is dictated by the diffraction limit. For many mesoscale scientific problems, it will be necessary to resolve far smaller detail than the micron length scale probed by typical IR microscopes. Therefore, we have pioneered the coupling of a broadband high-brightness synchrotron IR beam to the tip of an atomic force microscope (AFM) to enable full FTIR spectral fingerprints to be obtained with a spatial resolution only limited by the physical size of the AFM tip. Obtaining rich chemical spectra from 10-nm-scale regions will truly revolutionize the depth of information obtainable for molecular interactions and materials properties on the mesoscale.

Higher-resolution hard x-ray tomography. The ALS hard x-ray tomography beamline provides submicron-resolution tomographs that are often a useful complement to other higher-resolution images [e.g., Fig. 1(a)]. We are planning an upgrade of this beamline that will allow insertion of focusing optics, thereby providing ~100-nm resolution in 3D.

E. Emerging Mesoscale Analog Processors: From Catalytic Networks to Neural Processors

An important future goal of many of the research areas described in this strategic plan is to design and optimize mesoscale networks that integrate and connect nanoscale entities to achieve useful and often unusual functionality. For example, the "catalytic network" in Fig. 14 would be a micron-scaled reactor motivated by a biochemical reactor—a living cell. The various nanocatalytic centers would be connected with smart permeable membranes and allosteric regulators to optimize a particular function, in this case, low-temperature synthesis of useful fuels from small molecules with high efficiency and selectivity.

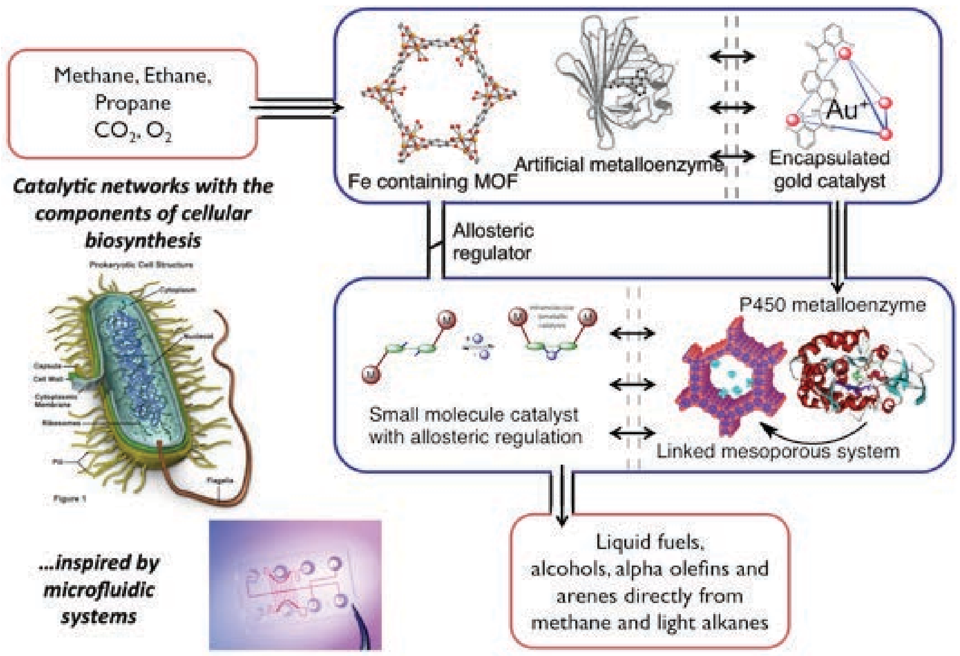

Fig. 14. Schematic of a possible catalytic network designed to take small feedstock molecules and produce complex fuels and other useful products, with high efficiency and selectivity at low temperature (courtesy of John Hartwig, UC Berkeley).

One can imagine uses for such networks in many diverse applications. A sensor network might mimic the sensitivity and specificity of a dog's nose, for example, that has more than 10,000 sensory neurons. The diverse phase behaviors and sensitivity to applied fields of multifunctional transition-metal oxides have already been proposed for many applications, and wiring them together in a network will be a logical next step to produce a functional mesoscale network, perhaps even to mimic neural processing. A precursor of that is emerging memristor technology that is proposed for neural processing. All such functional mesoscale networks can be considered to be analog processors that are optimized to solve highly nonlinear problems that are not well suited for digital computation and modeling. Neural processing is the classic example of such an analog processor.

These hypothetical devices would be composed of nanostructures that are already commonly measured using ALS tools—the metal-organic framework compounds, enzymes, porous membranes, and nanoparticles in the above catalytic network, for example. In such a functional network, the nanostructures would need to be probed selectively in situ and in operando to optimize the overall network. The spatial, chemical, and magnetic sensitivity of soft x-ray spectromicroscopy are well suited to address this important challenge. Several of the strategic beamline and endstation priorities in Table 1 will be of key importance as this work progresses:

- MAESTRO includes the nanoARPES endstation, which will measure the electronic structure of devices with a resolution of tens of nanometers. This includes plans to measure gated structures, which is likely how complex oxides, graphene, and topological insulators will be manipulated in a functional device.

- STXM is already used to measure simple functional devices—catalysts and batteries, for example. This will be a key capability of the AMBER beamline, which will have a STXM branch coupled to extensive sample preparation and complementary measurement capabilities that will be required to optimize a functional network.

- The COSMIC imaging branch will have the ability to probe the structural, chemical, and in some cases the magnetic structure of nanostructures in 3D, initially with ~10 nm resolution and ultimately, on an upgraded ALS (Sec. III.B), with few nm resolution. This is precisely the kind of capability one will need to map highly heterogeneous mesoscale devices, where complex interfaces and interphases will play a major role in device operation.

- Many mesoscale networks will involve coupled chemical reactions and reaction-diffusion processes. The COSMIC scattering branch, and more generally the suite of tools to measure nanokinetics, will be invaluable in probing these very complex processes at the nanoscale.

This section has been placed at the end of this document to provide a long-range focus for the ALS program—to show where the facility and its strong user community is likely to have a major impact in the coming decades. The ALS is very much on a path to have that impact, especially with the lattice upgrade described in Sec. III.B. It will be an exciting adventure to participate in this future of complex functional mesoscale networks.

III. Accelerator Renewal and Upgrades to Maintain World Leadership in Soft X-Ray Science and Technology

The ALS produces light for users over a wide spectral range, from far infrared (IR) to hard x-rays, with the core spectral region in the ultraviolet (UV) and soft x-ray region. In this core region, relevant to chemistry, catalysis, surface science, nanoscience, life sciences, and complex materials, the ALS remains competitive with the newest synchrotron radiation sources worldwide. The quality of the science program is directly connected to the performance of the accelerator complex and therefore continued upgrades of the accelerator have always been a high-priority activity for the ALS.

A. Upgrades Recently Completed or Near Completion

Brightness upgrade. A recently completed upgrade project improved the brightness of the ALS by reducing the horizontal emittance from 6.3 to 2.2 nm. This resulted in a brightness increase of a factor of 3 for bend-magnet beamlines and at least a factor of 2 for insertion-device beamlines. With this upgrade, the ALS has one of the smallest horizontal emittances of all operating third-generation light sources.

Controls/instrumentation upgrade. The controls and instrumentation upgrade is a four-year project that is scheduled to be completed in FY15. Its goal is to replace all outdated control system hardware and software, as well as much of the beam diagnostics hardware. This will enable the ALS accelerator staff to maintain and improve the reliability of accelerator operations, reduce the effort necessary to support the control system in the future, and provide improvements in performance, particularly in orbit stability. With the 20x improved bandwidth of the new BPMs and existing corrector, we expect a 2x improvement in the fast orbit feedback system.

Storage ring rf upgrade. The existing high-power rf system was nearing the end of its useful life and spares were becoming increasingly unavailable. Therefore an upgrade project is nearing completion with the goals of long-term maintainability, higher reliability, lower electricity consumption, and sufficient power reserves for all planned additions of new undulators, better immunity to AC line transients, and better fast phase stability. The main risk factor of the system, the old klystron, was replaced in FY12 and the project is planned to complete in FY15.

Major storage ring power supply replacement. The original large power supplies used for the four major magnet chains in the ALS lattice had become unreliable. In addition, newer designs can be more power efficient and provide better stability, thereby improving orbit stability. The last of the major power supplies was replaced in FY13.

B. Near-Term Upgrades

After the previously listed major upgrades are completed, many failure risks due to aging equipment will be retired. The largest remaining component is the injector rf system. In addition to the injector rf there will be need in the future to upgrade the 20-year-old single magnet power supplies for the 48 QF and QD magnets.

Injector rf upgrade. The rf systems in the ALS injector, with the exception of the booster rf amplifier, are all original to the ALS from the early 1990s. Due to their age, many of these systems, and in particular their components, are reaching the end of their serviceable life. In some cases, replacement parts are only available from single sources or are custom builds. While we have not experienced significant long-term outages of the system, there have been many shorter outages, and the rf group spends a significant amount of time maintaining the equipment. It is important to reduce the risk of prolonged beam outages caused by an inability to repair and maintain these systems and components. This includes the two 24-MW-based modulators and trigger thyratron, the amplifiers for the subharmonic bunchers, the master oscillator and distribution system, and the low-level rf controllers. Upgrading these systems would not only reduce this risk but give us an opportunity to increase performance and reliability.

In addition, there are ongoing infrastructure upgrade projects, namely efforts to reduce the frequency of beam losses due to AC line voltage fluctuations, to improve the temperature stability of the air in the endstation area, and to further improve water temperature stability. Several ongoing upgrades are also aiming at improving the overall energy efficiency of the ALS, both at the level of technical systems (rf, power supplies, injector operation modes), as well as building systems (HVAC).

Most of the insertion devices considered for ALS renewal are soft x-ray elliptically polarizing undulator (EPU) insertion devices. Other devices could be short-period undulators for harder (12-keV) photons. EPUs are very popular because of their great versatility in polarization and energy range. Along with other synchrotron facilities and exploring synergies with studies geared toward the Linac Coherent Light Source, we should explore possible avenues for improvement such as the following:

- non-mechanically-moving EPUs and/or improved field shapes,
- in-vacuum EPUs,
- cryogenically cooled permanent-magnet undulators, or
- Nb_3Sn-based superconducting undulators for achieving ultimate brightness in the soft x-ray range and for significantly higher brightness than is now achievable with wiggler sources for operation at 12 keV.

Pseudo-single-bunch (PSB) operation—a new operational mode at the ALS—can expand the capabilities of synchrotron light sources to carry out dynamics and time-of-flight experiments. In

PSB operation, a single electron bunch is displaced transversely from the other electron bunches using a short-pulse, high-repetition-rate kicker magnet. Experiments that require light emitted only from a single bunch can stop the light emitted from the other bunches using a collimator. Other beamlines will only see a small reduction in flux due to the displaced bunch. As a result, PSB expands the facility's capability to do timing experiments during multibunch mode. Furthermore, the time spacing of PSB pulses can be adjusted from milliseconds to microseconds with a novel "kick-and-cancel" scheme, which can significantly alleviate the complications of using high-power choppers and substantially reduce the rate of sample damage. On January 28, 2014, we introduced PSB into user time and the first experiments are being done with the kicker set at 4 kHz.

Beyond the baseline of the almost-completed brightness upgrade project, work is also investigating other possible modes of operation, including low-alpha modes, which are enabled by the fact that the new sextupoles allow control of the second-order momentum compaction factor. This could enable substantial improvements for THz experiments in special operation modes. To support these experiments, we are also considering modifying a vacuum chamber to enable larger acceptance angles for long-wavelength IR radiation. Multiparameter simultaneous optimization of the linear and nonlinear lattice with genetic algorithms is used as a tool.

Presently our vertical beam size in the insertion-device straight sections is just under 10 microns and our short-term orbit stability is about 0.6 micron (or ~6%). We are planning to reduce the coupling further and are adding a number of additional EPUs. At that point, the relative orbit stability would become significantly worse. The new BPM system will allow much greater orbit stability; however, to take full advantage requires a number (~20) of "fast" correctors—most likely air-core magnets surrounding bellow shields and associated power supplies. Complementing the improvements to fast orbit stability with the ongoing instrumentation upgrades, we are also considering improving the long-term pointing stability of our photon beams further by supplementing the photon beam diagnostics in beamlines. Also, the new storage ring timing system can allow the distribution of more precise timing signals to those users that require it.

Enabling insertion device(s) in Sector 2. There are 12 straight sections in the ALS. Currently 9 of these have insertion devices in them. The remaining 3—Sectors 1, 2, and 3—are presently being occupied by the injection systems (1), rf and multibunch feedback kickers (3), and the camshaft kicker and other systems (2). We are currently investigating the possibility of freeing up space in this sector to install one or two insertion devices while retaining its essential functionality for the storage ring. Among the ideas being considered are more compact multibunch feedback systems, replacing the four injection bumps with a single multibunch kicker, and relocating equipment. In addition to freeing up space, the multibunch kicker should greatly reduce injection transients, making top-off more transparent.

C. Longer-Term Upgrades

Evolutionary increases in storage ring source brightness over the past several decades like those described above have supported a robust array of x-ray capabilities that have had a major impact on many disciplines—physics, chemistry, biology, materials science, and others. A large capacity has been developed around the world and is applied to diverse, cutting-edge research problems. In recent years an additional, revolutionary increase in storage ring brightness has been proposed and is now being planned or pursued at facilities around the world. This increase will be accomplished by deploying storage ring lattice designs with electron-beam emittance comparable to the diffraction limit of the x-rays that are produced. That is, the x-ray beams will be nearly diffraction limited, with smooth, transversely coherent wavefronts. Coherence means that all photons are "useful" in demanding experiments that require focusing the beam into a small spot or encoding a material's heterogeneity into a far-field speckle diffraction pattern. For this reason, a diffraction-limited storage ring (DLSR) is ideal for examining the nano- and meso-scale structure of materials, since they enable the highest possible spatial resolution coupled to broad temporal sensitivity and x-ray contrast mechanisms. Groundbreaking new applications to study heterogeneous materials and functional devices will be possible on a DLSR.

We have therefore started to explore various lattice concepts for an ALS lattice upgrade to a multibend achromat lattice that would provide diffraction-limited soft x-ray beams up to about 2 keV, with decreasing coherent fraction at higher energy. We are focusing in particular on the balance between transverse nonlinear dynamics, longitudinal dynamics, and collective effects in an integrated way. The studies are currently at a preconceptual design stage and include numerical and analytical physics studies together with technology evaluation in the areas of magnets (DC + pulsed), vacuum systems, and rf systems (main and harmonic). To continue to serve the complementary hard x-ray community that coexists at the ALS, we are also studying concepts for advanced radiation-producing devices to incorporate intermediate x-ray sources into the lattice. An integral part is the optimization of possible injection schemes to enable high-brightness lattices by on-axis injection. The study of possible lattice choices and accelerator technologies will be informed by parallel efforts to further develop the science case for soft x-ray diffraction-limited light sources.

The goal of these design studies is to develop a firm proposal for an upgrade project in the existing ALS building and tunnel, keeping the same symmetry, location, and length of all insertion-device straight sections. The currently projected brightness envelope for this source is shown in Fig. 15.

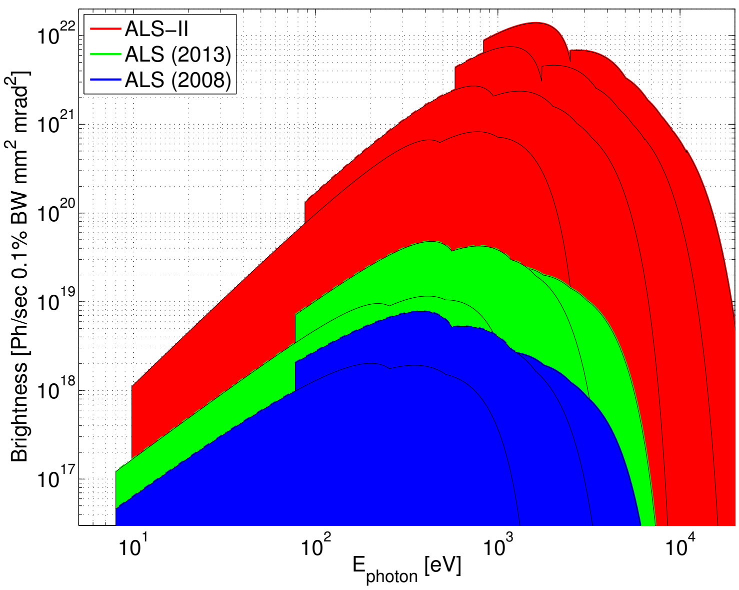

Fig. 15. Evolution of the brightness envelope of ALS undulators. Recent upgrades have improved soft x-ray brightness by about an order of magnitude, but upgrading to a multibend achromat lattice will provide a further increase by about a factor of 100.

Such an upgrade would endow the ALS with the highest soft x-ray brightness and coherent flux of any storage-ring-based x-ray facility planned or under construction. It would manifestly increase our users' ability to probe heterogeneous systems in ever-finer detail. It is a cost-effective upgrade that would allow a 20-year-old facility to maintain world leadership in soft x-ray science and technology for at least another 20 years.

IV. Appendices

A. List of Acronyms

AC: alternating current
AFM: atomic-force microscope/microscopy
ALS: Advanced Light Source
AMBER: Advanced Materials Beamline for Energy Research
AP: ALS Approved Program
APPES: ambient-pressure photoelectron spectroscopy
APS: Advanced Photon Source
APXPS: ambient-pressure x-ray photoelectron spectroscopy
ARPES: angle-resolved photoelectron spectroscopy
ARRA: American Recovery and Reinvestment Act
ASCR: Advanced Scientific Computing Research
BATT: LBNL Batteries for Advanced Transportation Technologies program
BCSB: Berkeley Center for Structural Biology
BER: DOE Biological and Environmental Research
BER/BSISB: BER-funded Berkeley Synchrotron Infrared Structural Biology program
BES: DOE Basic Energy Sciences
BPM: beam-position monitor
CCD: charge-coupled device
CMOS: Complementary metal-oxide semiconductor
COSMIC: Coherent Scattering and Microscopy beamline (ALS 7.0.1 complex)
CSGB: DOE Chemical Sciences, Geosciences, & Biosciences
CSD: LBNL Chemical Sciences Division
CRD: LBNL Computational Research Division
CXRO: LBNL Center for X-Ray Optics
DC: direct current
DLSR: diffraction-limited storage ring
DMSE: BES Division of Materials Science and Engineering
DOE: U.S. Department of Energy
EFRC: Energy Frontier Research Center
EHS: Environment/Health/Safety
EPU: elliptically polarizing undulator
ESAF: Experiment Safety Assessment Form
ESD: LBNL Earth Sciences Division
ESNet: Energy Sciences Network
EUV: extreme ultraviolet lithography
FEL: free-electron laser
FTIR: Fourier transform infrared spectroscopy
FY: fiscal year
GEMINI: new protein crystallography beamline
GPU: graphics processing unit
GU: ALS general user
HHMI: Howard Hughes Medical Institute
HVAC: heating, ventilation, and air conditioning
IR: infrared
JCAP: Joint Center for Artifical Photosynthesis, a DOE Energy Hub
JCESR: Joint Center for Energy Storage Research, a DOE Energy Hub
LBNL: Lawrence Berkeley National Laboratory

LCLS: Linac Coherent Light Source
LDRD: LBNL Laboratory Directed Research and Development
LUXOR: Lightsource Upgrade for X-Ray Optics Renewal
MAESTRO: Microscopic and Electronic Structure Observatory (ALS 7.0.2 complex)
MF: LBNL Molecular Foundry
MOF: metal-organic framework
MSD: LBNL Material Sciences Division
NA: numerical aperture
NCEM: National Center for Electron Microscopy in the LBNL Molecular Foundry
NERSC: LBNL National Energy Research Supercomputing Center
NSLS: National Synchrotron Light Source
PBD: LBNL Physical Biosciences Division
PEEM: photoemission electron microscopy
PNNL: Pacific Northwest National Laboratoy
PSB: pseudo single bunch
PSP: ALS Proposal Study Panel
QERLIN: Q- and Energy-Resolved Inelastic Scattering beamline
RAPIDD: Rapid Access, Industrial, and Director's Discretionary beam time proposal
rf: radiofrequency
RIXS: resonant inelastic x-ray scattering
RSoXS: resonant soft x-ray scattering
SAC: ALS Scientific Advisory Committee
SAXS: small-angle x-ray scattering
SB: superbend beamline
SEM: scanning electron microscope/microscopy
SEMATECH: Semiconductor Manufacturing Technology consortium
SIBYLS: LBNL Structurally Integrated Biology for the Life Sciences program
SISGR: DOE Single-Investigator and Small-Group Research program (2009)
spinARPES: spin-resolved ARPES
STXM: scanning transmission x-ray microscope/microscopy
SUFD: BES Scientific User Facility Division
SUFD/NSLS: SUFD funding to help with NSLS dark age
SUT: surface under test
SXE: soft x-ray emission spectroscopy
SXR: soft x-ray
TI: topological insulator
UC: University of California
UEC: Users' Executive Committee
USUP: User Services User Portal
UV: ultraviolet
VLS: variable line spacing (grating)
WAXS: wide-angle x-ray scattering
XAS: x-ray absorption spectroscopy
XFEL: European X-Ray Free-Electron Laser
XM-1: X-Ray Microscope #1 (ALS Beamline 6.1.2)
XPCS: x-ray photon correlation spectroscopy

B. Safe Operation on the ALS Experiment Floor

As part of Integrated Safety Management, the ALS Safety Program continuously evaluates the effectiveness of its program and identifies opportunities to improve. These improvements are integrated with, and support, the ALS Strategic Plan.

Integrating floor and accelerator operations. Several years ago, the ALS started cross-training of floor operators, who directly work with users and who play a key role assuring safety on the floor, and accelerator operators, who would normally have limited interaction with users and floor safety. This leads to better integration of these two activities and provides more staff trained to understand safety procedures and to notice potential hazards on the floor. Similarly, there are more staff who understand safe accelerator operations. This leads to a better and more efficient operation overall.

Management walk-arounds. ALS management schedules regular walk-arounds to understand floor operations, with a focus on safety.

Beamline/accelerator renewals and upgrades. The scope and nature of both beamline and accelerator projects is constantly evolving, and the EHS review processes need to evolve with them. Many projects involve upgrades of current systems and so “abbreviated” review processes are being developed that focus resources more effectively. Also, operating experience with safety management systems such as top-off critical apertures, beamline shielding endpoints, beamline radiation safety training, and other issue areas is being used to make them more effective and efficient. Lastly, current DOE trends in accelerator safety, including unreviewed safety issues, configuration control, readiness reviews, etc. are being integrated into ALS processes.

Technical capabilities. The ALS Safety Program is collaborating with other elements of the ALS in a number of areas: developing a strong user biology lab and technical support function; continuing development of chemical lab support such as glove boxes; mezzanine lab capabilities; in situ capabilities for flame, low/high temperature, high pressure, laser, and other potentially hazardous environments; and improved support in the handling of low-level radioactive materials.

Scientific support. As discussed in more detail in Appendix D, a primary goal of developing a new User Portal is to develop a simple and intuitive web-based input tool for users to create safety analysis forms for each of their experiments. This is being modeled on the APS system and will strive to use the same terminology and have the same outputs (controls) for similar experiments. Much of this is being piloted in the current RAPIDD project.

C. ALS Workforce Development

The success of the DOE synchrotron radiation facilities depends strongly on developing a knowledgeable and highly trained community of users and beamline scientists who apply existing tools and innovate new tools, often collaboratively, to pursue a diverse array of research frontiers. The ALS takes its role in workforce development very seriously.

The ALS takes very seriously the need to seek internal and external recognition for its scientific staff, particularly the beamline scientists who are at the heart of our success. We have regular successes on internal ALS, LBNL, and DOE awards, and we have had recent successes with external recognition as well:

- In the past year Howard Padmore was awarded the AVS Albert Nerkin Award and Fernando Sannibale was elected Fellow of the APS.
- Staff are regular recipients of annual ALS awards for excellence in research, instrumentation, and service—the Shirley, Halbach, and Renner Awards. These are awarded during the annual User Meeting and selections are made by the ALS Users' Executive Committee.
- With several excellent staff who have been at the ALS almost since the facility was commissioned, the ALS Division Staff Committee has placed renewed emphasis on nominating more ALS staff for promotion to LBNL Senior Staff Scientist, the rough equivalent of a full professor at a university. We achieved one such promotion last year, and are working on two such promotion cases this winter.
- ALS management focuses on developing strong candidates for DOE Early Career Awards. Our first success occurred in the past year with Alex Hexemer. A key feature of success in this was simply nominating an exceptionally strong candidate who is also an exceptionally strong, collaborative, and productive beamline scientist. We have more young candidates who fit that mold and who are nominated this year. We will continue to hone our process in the future.

External recognition for beamline staff is a more difficult problem since the portfolio of a beamline scientist does not map very well onto the criteria for external awards, which tend to be heavily oriented toward individual research contributions. The collaborative style and heavy service load of our staff (and those of other user facilities, of course) is not well matched, for example, to society awards. But our users understand and fully appreciate the efforts of our staff: the fact that the UEC regularly recognizes them with internal ALS awards provides tangible evidence for that.

As discussed above, our strongest beamline scientists establish very strong records of collaborative research, to the extent that they are equal partners with users in our strongest research activities. Postdocs with external fellowships now seek to come to the ALS to work with these strong staff members in these collaborations. This is the kind of activity that allows them to establish the credentials needed to be considered for external awards. The ALS has established an awards committee composed of the Division Deputy for Science and the leaders of the Scientific Support Group and the Experimental Systems Group, with others as needed, to focus on organizing nominations for these awards in timely fashion. We will nominate three or four of our staff for society fellowships this year. Such fellowships are an important first step, and the committee will be continuing to organize other nominations in the future.

Also important is to establish a strong pipeline of talented candidates to become facility staff in the future. Aside from the user training activities that happen daily on the experiment floor, the facility sponsors three related programs that directly impact the professional development of young scientists, from college undergraduates to advanced postdoctoral associates:

- ALS Post-Baccalaureate and Internship Program: The ALS allocates $375K/year to support undergraduate interns and recent college graduates for part- or full-time employment at the facility for a period up to one year. These students are assigned to work closely with an ALS staff scientist and to help with a project that will update or expand the capabilities of an ALS beamline or endstation. Alumni of this program are competitive in applying to the best graduate programs, and regularly select an ALS user as their doctoral thesis advisor. Other alumni have been hired for technical support positions at the ALS, elsewhere at LBNL, or in the high-technology industry.

- ALS Doctoral Fellowship Program: The ALS allocates $175K/year to this program, which supports typically eight to nine doctoral fellows in steady state. This program is highly competitive and attracts superb young talent to the ALS. The ALS offers each Fellow support of about 50% of a typical graduate student's pay. The Fellow's thesis supervisor generally provides the balance of financial support as well as university benefits. In addition to training, other goals of the program are to engage the thesis advisor deeply in ALS research activities and to provide a career development opportunity and supervisory responsibility to ALS staff scientists. The program was established in 2003 and has developed an impressive list of alumni.

- ALS Postdoctoral Fellowship Program: The ALS allocates $695K/year to this program, which fully or partially supports four to six postdocs in steady state, depending on leveraging. The strength of the ALS in applying x-rays to frontier research problems attracts a very strong pool of applicants. The financial arrangements are more diverse than for the doctoral program, but the funds are often similarly leveraged and a primary intent is again to engage faculty as well as PIs in other LBNL divisions in strong collaborations. The rapid development of ALS programs in energy science, for example, has greatly benefited from strategic allocation of these funds in collaboration with principal investigators in the LBNL Environmental Energy Technologies Division. About one-third of the alumni of this program are hired through a regular search process as beamline scientists at the ALS or at other x-ray facilities, and a similar number are hired into faculty positions around the world and now direct their own research programs using synchrotron radiation.

There is a sizable flux of students from one of these programs into the next: a few interns and post-baccs become doctoral fellows, and several doctoral fellows become ALS postdocs, either in the fellowship program or supported by an LDRD proposal. Given this training, it is to be expected that many of the postdoctoral fellows continue their careers as beamline scientists or active users of synchrotron radiation facilities. These workforce development programs have a significant long-range impact on the synchrotron radiation community at large and on the ALS user program specifically, even though fellows are not allowed to engage directly in user service.

The programs have been approved and supported by the DOE and are part of the ALS Field Work Proposal. The fellowship programs qualify for a significantly reduced LBNL indirect cost rate, which, combined with the leveraging described above, makes them very cost effective. The doctoral and postdoctoral programs are apparently unique among the DOE light sources. They are discussed with the ALS Scientific Advisory Committee and always receive very positive reviews.

Committees formed from the ALS staff and user community oversee these programs and help maintain a high degree of leveraging and ensure strong alignment of the programs with the facility's strategic plan. Applications for doctoral fellowships are solicited annually, and files are evaluated by an ad hoc committee composed of the ALS Division Deputy for Science, the head of the ALS Scientific Support Group, a beamline scientist, and usually two faculty who conduct strong research programs at the ALS. A separate committee composed of the Division Deputy for Science, ALS group leaders, and two beamline scientists meets regularly to consider ALS needs for postdoctoral staffing and to evaluate applications. More detailed information about the programs, including several "postdoc highlights," is provided on the ALS website at

http://www-als.lbl.gov/index.php/component/content/article/56-general-resources/405-als-doctoral-fellowship-in-residence.html

http://www-als.lbl.gov/index.php/component/content/article/56-general-resources/401-als-postdoctoral-fellowship-program.html

http://www-als.lbl.gov/index.php/resources/employment/780-als-postdoctoral-fellowship-highlights.html

These workforce development programs are strongly coupled to the model the ALS uses to achieve its highly successful operation. A key ingredient of that success is career development of our beamline scientists, who through their collaborative participation in cutting-edge research are highly motivated to develop new instruments and capabilities and to build a strong user program. Some of the most successful ALS beamline scientists have learned how to leverage resources available to them, including their own small beam-time allocation, beam time of their close collaborators, funds from these fellowship programs, and other resources brought by their collaborators and provided by the ALS. The fellowship programs are a crucial ingredient of this process, both in supporting existing beamline scientists and training new ones.

D. Developing a Modern User Portal

The ALS User Services web interface is the first face of the ALS seen by ALS users. Provision of a modern User Services business system, for coordination of user registration, proposal administration, safety management, and tracking and reporting outcomes, is essential to ensure safe and efficient user operations at ALS. The last review of the ALS by BES recognized that a more integrated web-based system was needed, and that the ALS should consider ways to reduce the lead time for general user access. To this end, the ALS has initiated a project to implement a web-based User Portal to provide a technologically relevant strategic systems platform that will mitigate risks associated with the current platform and meet ALS needs for at least the next decade with an easy path to future requirements. The new User Services User Portal (USUP) will provide a single point of entry to a personalized, easy-to-use interface that will allow users to track past, current, and future beam-time applications and complete all safety and security requirements.

ALS has planned a step-by-step approach to the USUP development that will provide useful tools at each step. The first step, a functional analysis of the business needs, has been completed and the requirements of User Services can be summarized in nine key processes (Fig. 16). Three of these processes have been identified as near-term priorities for the USUP and are outlined in red. Our current capability in these three areas handicaps the provision of a quality service to our users, as well as creating increased workload for both users and staff.

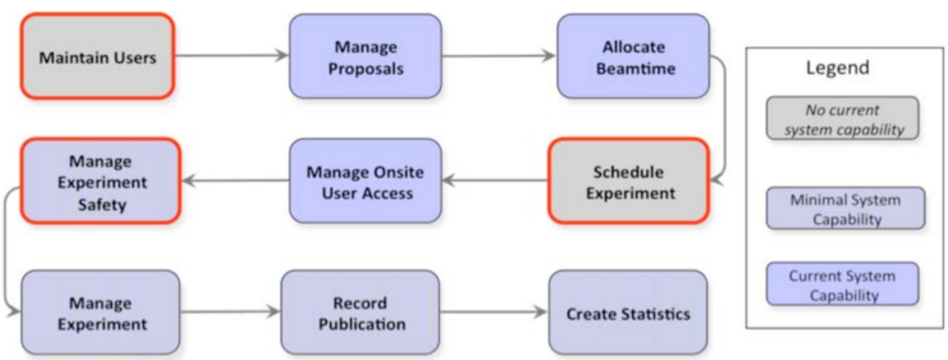

Fig. 16. Nine key processes summarize the requirements of User Services.

Phase one of development started in 2012 and involves implementation of the three priority processes described below.

Manage experiment safety. Ensuring a safe experiment environment is critical to ALS success, and an upgrade of the user safety process is the highest priority of the USUP. The process utilizes the Integrated Safety Management framework to identify and control hazards associated with the tools, materials, procedures, and persons participating in ALS experiments. Design of the new system is at an early stage and is influenced by peer systems at other U.S. light sources.

Each experiment is required to provide information about samples and potential hazards through an Experiment Safety Assessment Form (ESAF) created in the system. Repetitive experiments may replicate a previous ESAF to streamline the process, updating only the entries that differ for the upcoming experiment. Online tools will ensure that creating, completing, and reviewing the ESAF will be an integrated process. Hazard identification is a critical component of any safety system, and the new system will employ sophisticated decision making to support users and reviewers. Training, including, on-the-job training, will be integrated into the process.

Maintain system users and roles. This process is a requirement before implementing all the other processes and, for each user of the USUP, establishes the roles of that person. For all system users, the relationship with the ALS begins with self-registration and collection of basic information. Once registered, system users can manage their personal information and may participate in the appropriate processes for their defined roles. The eight roles that have been identified for system users and the extent of involvement of each role can be described by mapping the roles onto the nine key processes (Fig. 17).

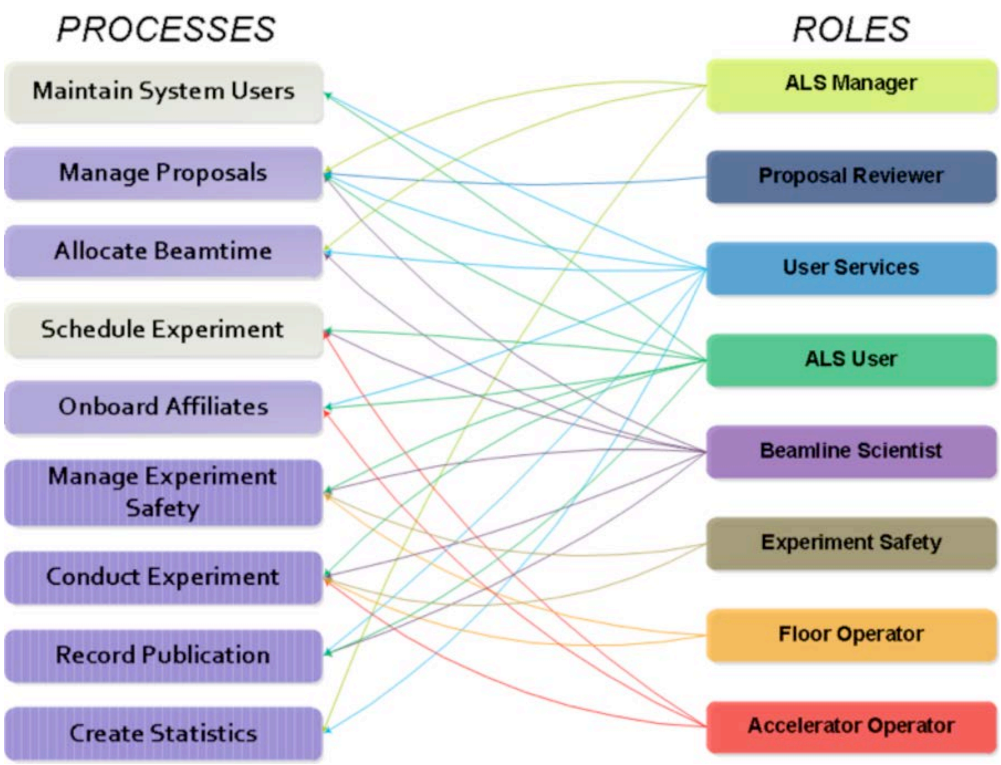

Fig. 17. Mapping of roles into processes.

Schedule experiments. A centralized scheduling tool is a prerequisite for efficient flow of data from proposals through experiment safety through reporting, and should increase staff productivity. The ALS has no current centralized capability to schedule beam time; instead beamline schedules are developed independently at each beamline. This tool will provide data for ALS to manage experiment safety and will enable accurate tracking of beamline usage and optimize efficiency at each beamline. High-level requirements have been identified for this process and these are being used to assess the scheduler recently implemented at the Advanced Photon Source. Reuse of existing software within the DOE umbrella is likely to reduce the total cost of the development in addition to increasing the familiarity of the user experience.

An active proposal with beam time allocated is a prerequisite before beam time can be scheduled. The RAPIDD proposal process, described below, will ensure this need is met for all categories of facility access.

RAPIDD proposal process. The majority of ALS experimenters are allocated beam time through the biannual General User Proposal and Approved Program processes. An upgrade of the existing proposal system has been started to develop a flexible process that will generate proposals for access routes that are currently handled manually or by Participating Research Teams. The new process, called RAPIDD, also enables rapid-access peer-reviewed general user and industry access, and complements the existing biannual proposal processes.

Phase one development of USUP will be rolled out this spring. In addition, ALS is working with LNBL to ensure that a lab-wide system under development for recording scientific publications will be available to meet ALS needs. After completion of phase one, additional phases are planned to upgrade the ALS proposal system, the process for on-site user registration, and production of statistics and reports. Upgrading the User Services interface for ALS users is seen as critical to ensuring safe and efficient user operations.

E. Engaging the ALS User Community to Develop a Strategic Path to Excellence

This Strategic Plan is as diverse as the ALS user community because the ALS seeks regular advice from many different sources:

- We engage our Users' Executive Committee (UEC) formally and informally several times per year on a host of issues to discuss how we might help users be more productive at the facility. The UEC is elected from the user population and generally represents the spectrum of research activities at the ALS.
- The UEC also organizes our annual User Meeting, which includes typically 12–15 topical workshops organized collaboratively by our staff and users (see *http://www-als.lbl.gov/index.php/user-information/users-meeting/835-2013-workshops.html*). These workshops provide invaluable advice on emerging opportunities and research priorities. They are supported financially by the ALS—an indication of how valuable we think they are—and are very well attended.
- The ALS is now part of the LBNL Energy Sciences Area, the heart of DOE BES activity at the Laboratory. Regular Area Meetings with partner divisions provide valuable collaborative strategic planning. For example, the ALS helped guide the discussion of potential EFRC proposals from LBNL and partnered with researchers from across the Energy Sciences Area to develop these.
- The ALS has strong ties with many faculty and research groups from UC Berkeley and other UC campuses, and these also contribute to our strategic thinking.
- With oversight from our Scientific Advisory Committee (SAC), the ALS organizes two reviews per year of entire subdisciplines to seek focused advice on how to optimize our capabilities to address important research problems. In the past year we have held two such reviews, one in atomic and molecular physics and another in magnetism.
- The ALS SAC, composed of national and international experts from many different disciplines, meets twice per year to provide high-level advice on our program. The ALS has enjoyed and fruitful and productive interaction with its SAC for many years. All of our strategic plans and priorities are discussed in detail with the SAC.
- The Approved Program (AP) process is another key feature of the ALS planning process. AP proposals are evaluated by the Proposal Study Panel (PSP), which also evaluates regular general user (GU) proposals. AP proposals must describe a research program that is of very high quality evaluated against the GU population and must also propose to help the ALS develop its capabilities in some significant way. The final recommendation on an AP proposal is made by the SAC, which also gauges the research and development plans in terms of the overall ALS program.
- Finally, in a fashion similar to the Approved Programs, the ALS doctoral and postdoctoral programs described in Appendix C provide yet another channel to engage strong users and to leverage ALS resources toward future strategic priorities.

F. Advanced Detectors

The ALS Detector Development Group builds on LBNL's history of particle detectors, in particular the past three decades focused on highly integrated, microelectronic-enabled detectors. The group's focus is on the development of novel soft x-ray detectors that enhance the productivity of the ALS and enhance the facility's scientific reach. Workshops (every one to two years) at ALS User Meetings are forums to collect community need and interest, and, together with the Experimental Systems and Scientific Support Groups, drive our priorities. Research and development activities are currently funded under the BES Accelerator and Detector program and are used to initiate, design, and prototype novel detector concepts. We greatly benefit from ALS Beamline 5.3.1, dedicated to detector and optics testing, as a facility to characterize and validate detector concepts. ALS operating funds are used to deploy the detectors we develop and to adapt them to specific experiments and beamlines.

Recent activities have focused on direct-detection CCDs capable of capturing hundreds of megapixels per second, developed in collaboration with the Detector Group at APS. These have been (or are currently being) deployed at ALS, APS, LCLS, NSLS-II and XFEL. Current activities include the following:

- Development of CCD detectors 100x faster than those described above—currently in the prototype and evaluation phase.
- Development of very fine-pitch detectors for use in spectrographs. A 5-µm-pitch device has been produced and demonstrated and is of interest for photon-in/photon-out experiments worldwide.
- Development of very thin conductive entrance windows. Soft x-rays, at low energies, have very shallow penetration depths, so that extremely thin entrance contacts are required in order to avoid a precipitous loss in quantum efficiency. Further, to be used on fully fabricated semiconductor detectors, the contact process must be low-temperature (less than the melting point of the metal interconnects). We have developed a successful and simple process for 100-nm contacts and are performing research and development on contacts as thin as 10 nm.
- Development of monolithic soft x-ray CMOS detectors.

Future plans include the development of sensors with avalanche multiplication (for high signal-to-noise soft x-ray single-photon counting) and ultrafast readout based on advances in nanometer CMOS.

As faster detectors mean higher data rates, our activities are intimately connected with efforts on "big data," both the current, straightforward, efforts at high-bandwidth transport to high-performance computing centers, and future research and development toward in silico algorithms for on-detector data reduction.

G. Lightsource Upgrade for X-Ray Optics Renewal (LUXOR)

The cost of new beamlines at the ALS is typically between $4M—$8M. We are now at the stage where we are starting to replace the oldest beamlines at the ALS, designed over 20 years ago. During this time, the performance of the source has radically improved in terms of brightness and stability, x-ray optics vendors can produce optics to much higher specifications in more complex shapes, and optical designs have become much more sophisticated.

As well as seeking funding for the complete replacement of beamlines, a prudent, rapid, and very cost-effective approach to lengthening the lifetime of our existing complement of beamlines will be to invest in replacement optics, typically up to 10% of the replacement cost of the beamline. These replacement optics will be of higher quality and more sophisticated design and allow us to take advantage of the current higher brightness of the ALS, compared to its brightness when these beamlines were built. We will also be able to solve many of the outstanding issues of beamline performance, where optical components have aged and are no longer operating to specification.

We have a very clear demonstration of the effectiveness of this type of optics upgrade. Five years ago, in partnership with the Berkeley Center for Structural Biology, a program was initiated to upgrade optics in three of our structural biology beamlines. The performance of these systems had decreased over the years, but by judicious replacement of key optics, we were able to gain factors of up to 100 in flux onto a small sample.

We have examined what needs to be done on all BES-funded beamlines around the ALS to bring their performance up to specification and offer a performance commensurate with the new higher-brightness upgrade of the ALS. We find a range of benefits and solutions ranging from situations where no upgrade is required, on some of the newer beamlines, to a situation where replacement of most optics is required, on some of the older beamlines. In the latter case, improvements of photon flux on the sample can be up to two orders of magnitude or more.

The typical cost of upgrading a beamline's optics would be around $0.5M. We could do upgrades at a rate of three per year, for a cost of $1.5M/year. In total, we would upgrade around 15 beamlines over five years, for a total cost of $7.5M. Early targets of this program are listed below.

- Beamline 4.0: Replacement of plane pre-mirror and conversion to VLS design.
- Beamline 8.0: Replacement of pre-mirror, grating, and refocus optics.
- Beamline 10.0: Replacement of gratings and pre-mirror optics.
- Beamline 11.0: Replacement pre-mirror, elliptical refocus mirror, and branch mirrors.
- Beamline 9.3.2: Replacement of pre-mirror, gratings, and refocus optics.

We expect the upgrades above to increase resolution, throughput, or stability by at least an order of magnitude over current performance.

H. State-of-the-Art X-Ray Optics Metrology Laboratory

The ALS's ability to study inhomogeneous materials and material devices depends directly on the ability of beamline optics to transmit and focus with high fidelity the very bright ALS source onto a sample located in a complex environmental cell. Our collaboration with CXRO to develop diffractive optics, for example, is very productive and is essential to our long-term success. A new metrology lab is also an essential ingredient of testing and aligning grazing-incidence optics.

Construction of a new optical metrology laboratory in the ALS User Support Building, with comprehensive control of environmental conditions, was completed in summer 2013. Tests have shown that the lab is a cleanroom facility with a rating better than class 1000 and temperature stability better than ±30 mK over a day. In late November 2013, the laboratory began operation once the existing metrology instruments were moved from the old lab, upgraded, and recommissioned. Because the lab's capabilities now extend far beyond classical optical surface metrology to include the entire spectrum of in situ and ex situ metrology, and because it also supports the design and fabrication of x-ray optics and optical and mechanical systems, the new lab has been renamed, from the Optical Metrology Laboratory to the X-Ray Optics Laboratory.

The laboratory assures the quality of the optical components installed in beamlines or used for experimental systems. This entails measuring mirrors to ensure vendor compliance to specifications, calibrating and adjusting bending parameters for adjustable mirrors, and thorough alignment, tuning, and characterization of optomechanical systems. Usage of different instruments ex situ enables us to separately investigate and address different potential issues affecting the beamline performance of an optic. These are surface quality (figure and finish errors), temporal and temperature dependence of surface shape, mechanical stability, gravity effects, alignments (twist, roll-off, yaw error), etc. At the beamline, all the perturbations produce a cumulative effect on the beamline performance of the optic, making it difficult to optimize performance. Ex situ metrology allows us to fix the majority of the problems before installation of the optic at the beamline and to provide feedback on design and guidelines on usage (for example, ambient-temperature stability and accuracy of alignments).

The equipment utilized in the lab includes a phase-shift interferometry microscope, a large-aperture interferometer, two slope-measuring long-trace profilers (the LTP-II and DLTP), an atomic force microscope, optical microscopes, a differential laser Doppler vibrometer, and various systems for the development of new x-ray optics and metrology techniques. To fully realize the advantages of the new optics lab, a new high-precision granite gantry system, with air-bearing translation systems capable of precision 2D scanning over the surface under test (SUT) as well as tilting and flipping the SUT, has been specified and purchased (delivery is scheduled for the end of March 2014). The system is a key element of a new instrument under development for surface-slope metrology at levels below 50 nrad.

I. The Data Handling and Analysis Opportunity

The need to capture, transmit, store, and analyze large-volume data sets at DOE x-ray facilities and beyond has been well documented. The ALS's participation in this problem is confirmed by Fig. 18, which shows the amount of raw data collected at several ALS beamlines in the past, present, and future. In 2013, we projected that the facility will generate a total of ~2 petabytes of data. The problem is driven by a combination of high-brightness sources, efficient and highly parallel detectors, and increasing implementation of combinatoric and rapid screening protocols.

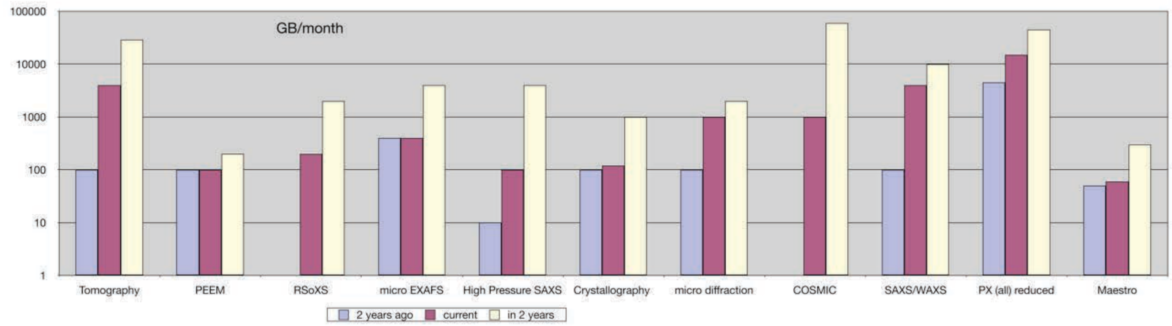

Fig. 18. Amount of raw data collected at ALS beamlines over the past three years.

Fig. 19 emphasizes the data analysis problem, which is likely the most complex part and the part that will take the most effort and time to solve. A single tomography beamline can presently produce ~10 Tb of data in a single day. Through a series of steps requiring complex software, this will be reduced to a publishable graph contained in a ~10 Mb image. The data handing capacity is being developed based on known technology, and very soon the efficiency of this process will be determined by data analysis software.

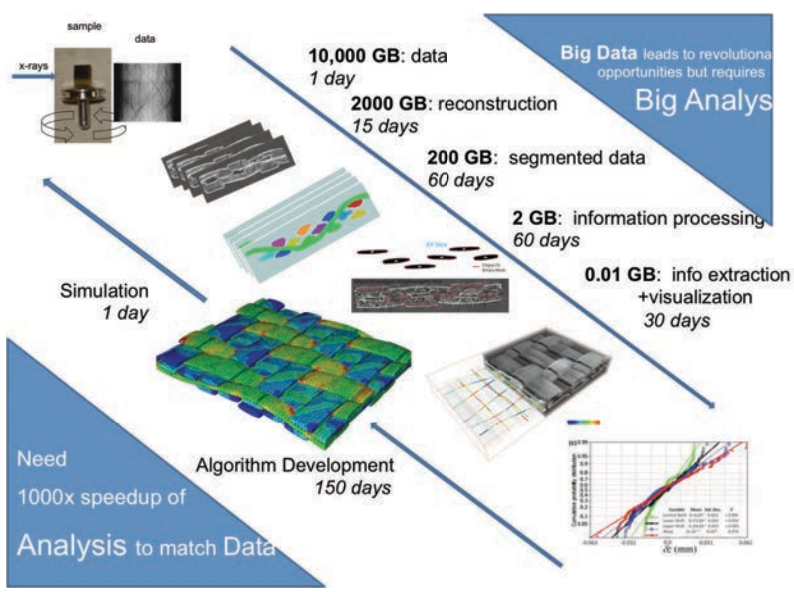

Fig. 19. The path to analyzing a large data set to a publishable figure requires several steps, all of which need efficient, user-friendly software that in many cases remains to be implemented.

The path to addressing the "big data" problem lies in a comprehensive system of data capture, management, analysis, and integration. At a user facility, these activities are set against a backdrop of collaboration in distributed environments. With the notable exception of particle physics, state-of-the-art data capture and management infrastructures for facilities are not widely applied, and high-performance data-analysis pipelines are rare. Few x-ray facilities operate data archives, and none integrate data within or across facilities and its users.

The ALS, in collaboration with the LBNL-based and ASCR-funded Energy Sciences Network (ESnet), the National Energy Research Scientific Computing center (NERSC), and LBNL's Computational Research Division (CRD), has been addressing this problem. The approach is

- to deploy advanced data-transfer techniques and to make these available to the ALS community (ESnet and NERSC),
- to provide hands-free data and metadata packaging and transfer from ALS beamlines to the NERSC High Performance Storage System, and
- to develop a suite of tools including (1) a robust analysis pipeline, framework, and services, and (2) on-demand data access, processing, and simulation (CRD).

Our efforts to date have focused primarily on the first two steps, in the belief that we need to establish a robust infrastructure before the third can be seriously addressed. A prototype data pipeline has been implemented on three high-data-rate beamlines that moves data seamlessly to NERSC and makes it available from a web browser. A fourth beamline (COSMIC imaging) is being developed with similar data management capacity. An initial suite of high-performance analysis codes is being developed in four areas served by these beamlines:

- X-ray microtomography: reconstruction code, reconstruction, image filtering, and segmentation.
- Grazing-incidence and transmission small-angle x-ray scattering: multi-GPU code for Reverse Monte Carlo.
- X-ray microdiffraction: XMAS microdiffraction analysis software.
- COSMIC imaging: real-time phase retrieval for ptychographic data sets, image processing.

In all of these cases, an important motivation is to increase the efficiency of ALS beamline usage by online and nearly real-time (preliminary) data analysis to provide users with guidance for what to do next. We also recognize that large data sets may be useful to other researchers beyond the data generator. ALS will evolve data policies consistent with those being developed by the Office of Science.